

# **Decadal multi-constituent chemical reanalysis and its applications in air quality and climate research**

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K. W. Bowman, D. Fu, J. Neu, G. Osterman, S. S. Kulawik, M. Lee, J. Worden, Z. Jiang, T. Sekiya, K. Sudo, Y. Kanaya, H. Eskes, K. F. Boersma, V. Huijnen, A. M. Thompson, B. Gaubert, J. Barre, L. K. Emmons, H. Worden, D. Henze, B. McDonald, T. He, D. Jones, A. Arellano, K. Yumimoto, T. Walker, J. Flemming, A. Inness

# Air Pollution May Be As Harmful To Your Lungs As Smoking Cigarettes, Study Finds



*Assessed nearly 7,000 adults living in six U.S. cities. Published in JAMA (IF=51.3)*

**An increase of about 3 ppbv ground-level ozone outside your home over 10 years in a low pollution area was equivalent to smoking a pack of cigarettes a day for 29 years.**

So, even moving from a low-pollution area into one of the cleaner cities could still increase your risk of respiratory diseases like emphysema.

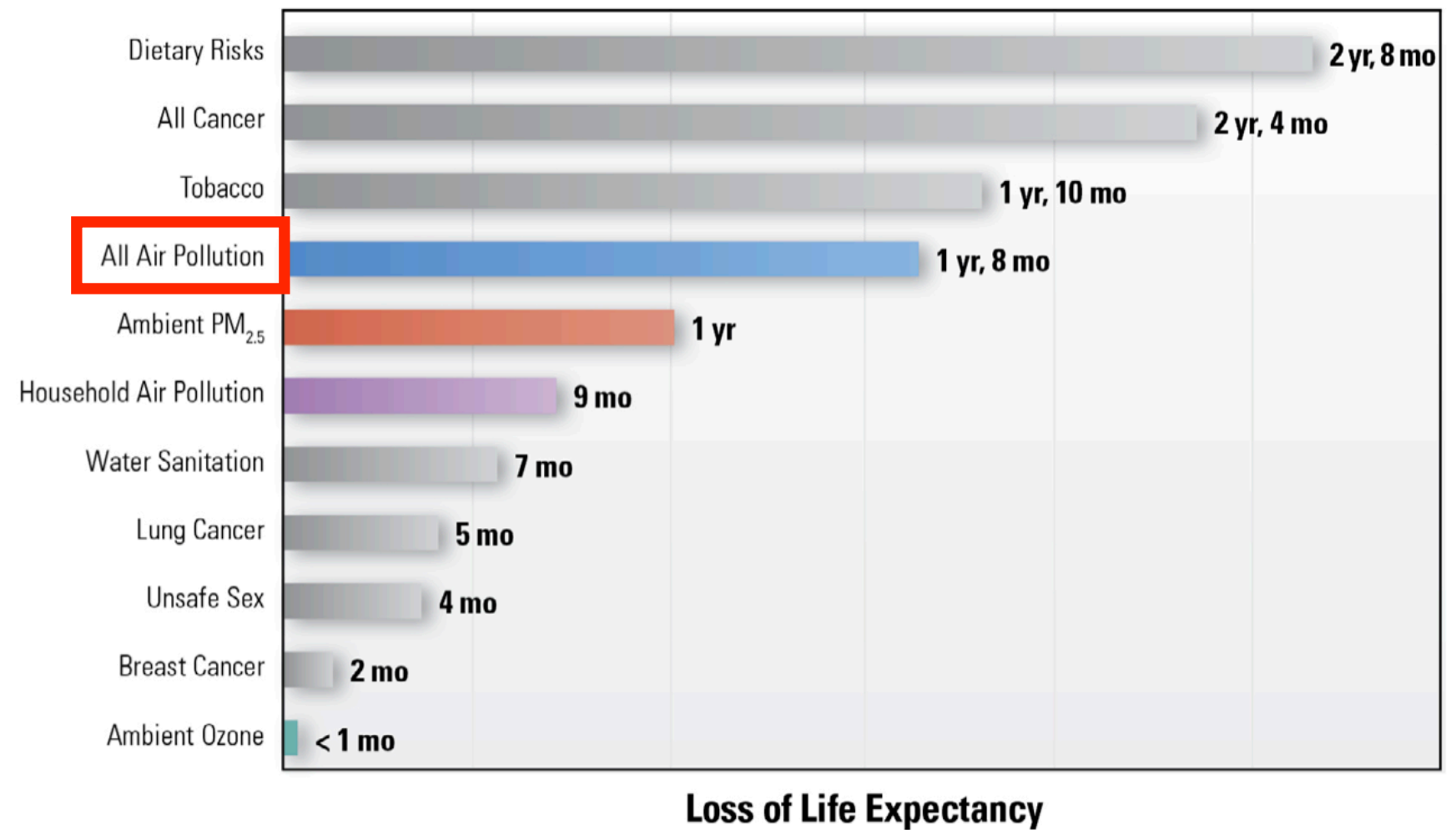
WHO calls urban pollution levels a “public health emergency”

- **91% of us live in areas where air pollution exceeds the agency’s guidelines.**
- **55% of the world’s population lives in urban areas, rise to 68% by 2050 (UN).**
- If urban life is a serious concern for our health, then it is only going to become a greater issue for more of us as time passes. (BBC)

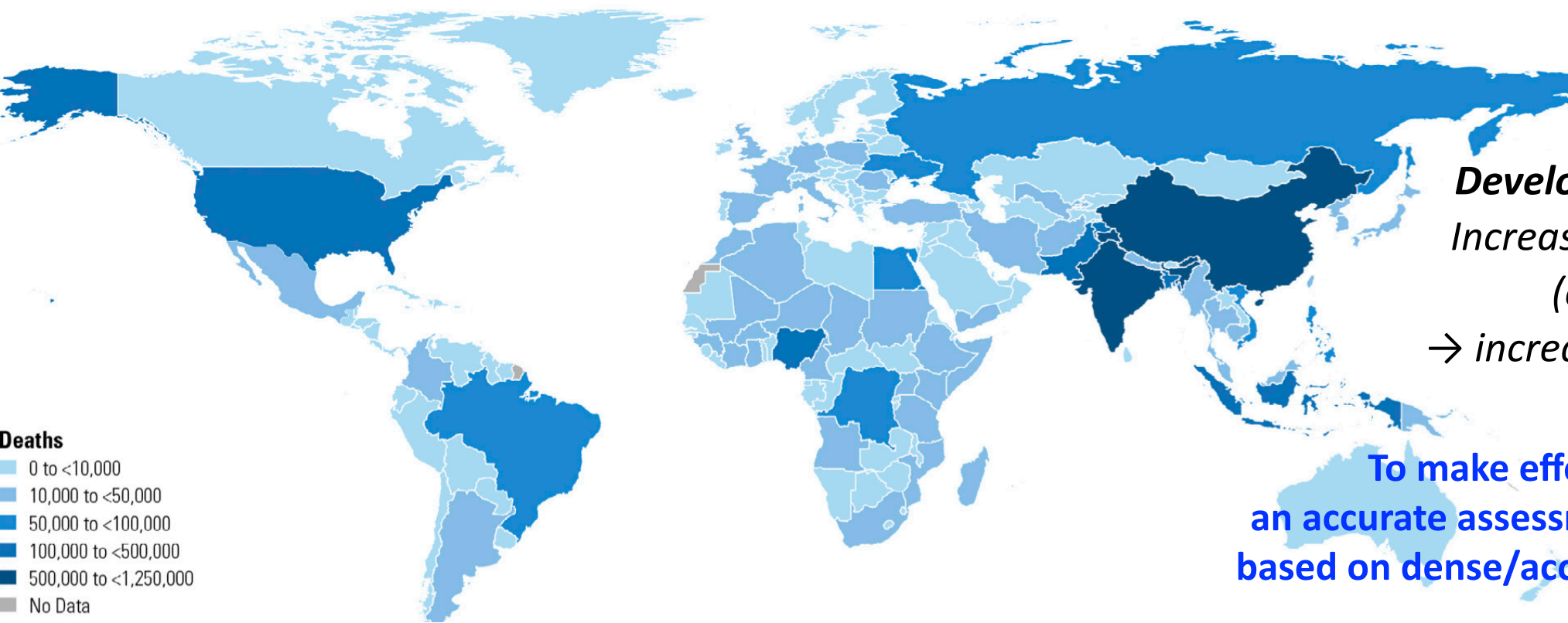


## Contribution of major risk factors to loss of life expectancy.

*(State of Global Air, 2019)*



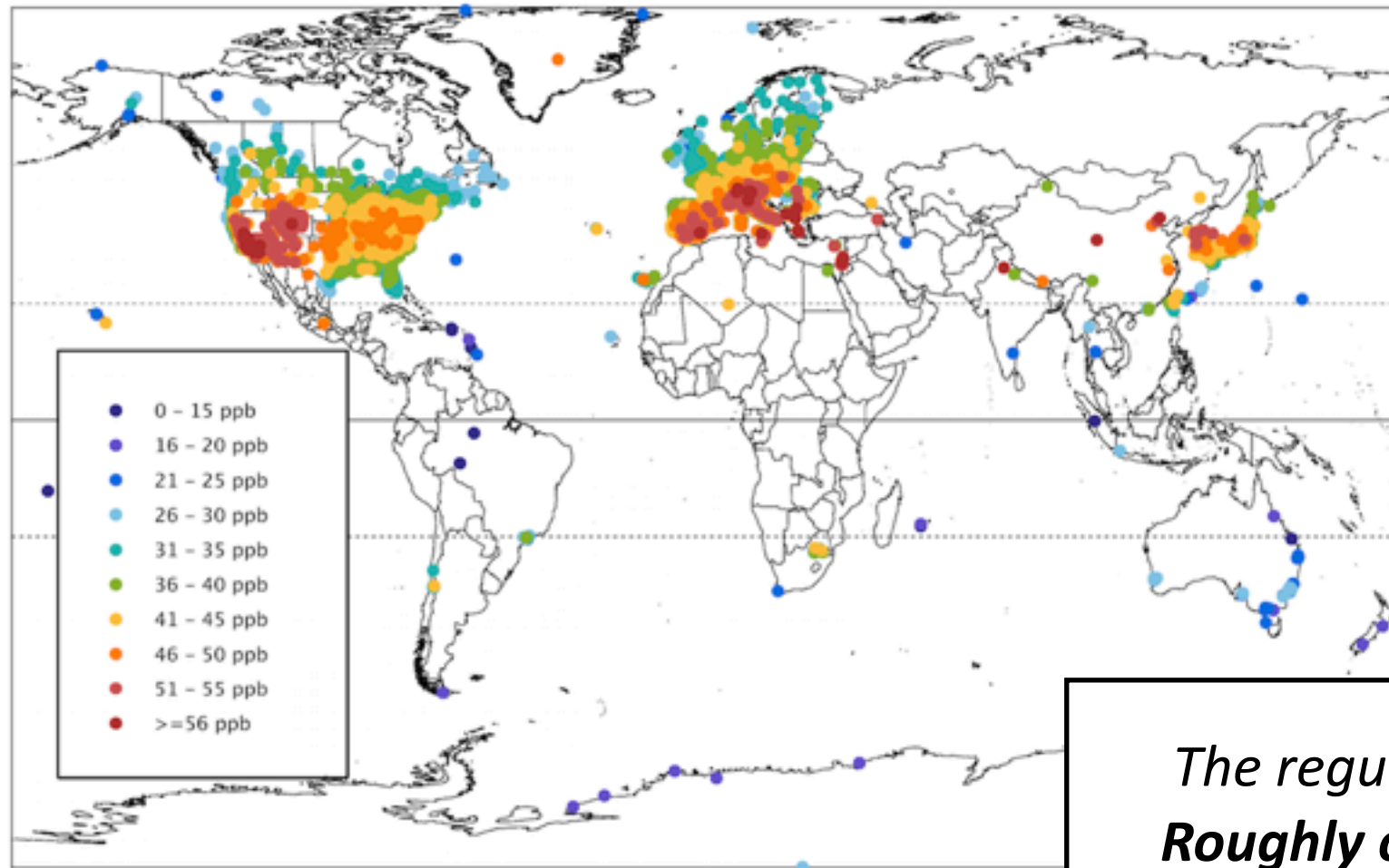
## Numbers of deaths attributable to air pollution in countries around the world in 2017.



*Developing countries in the tropics  
Increased emissions & stronger OPE  
(e.g., Zhang et al., 2016)  
→ increased air pollution exposure risk*

**To make effective policy guidelines,  
an accurate assessment of air pollutant exposure  
based on dense/accurate measurements is needed**

Daytime average, summer Data extracted on: 2016-10-24  
daytime-avg ozone, 2010-2014 (minimum 3 years): 4794 all sites



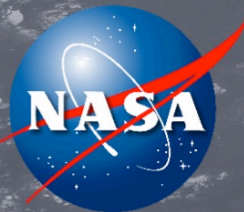
**TOAR**  
tropospheric  
ozone  
assessment  
report

*The regular surface ozone monitoring sites:  
Roughly only 17% of the global population!*

## For improved assessment of long-term changes in global air pollution exposure

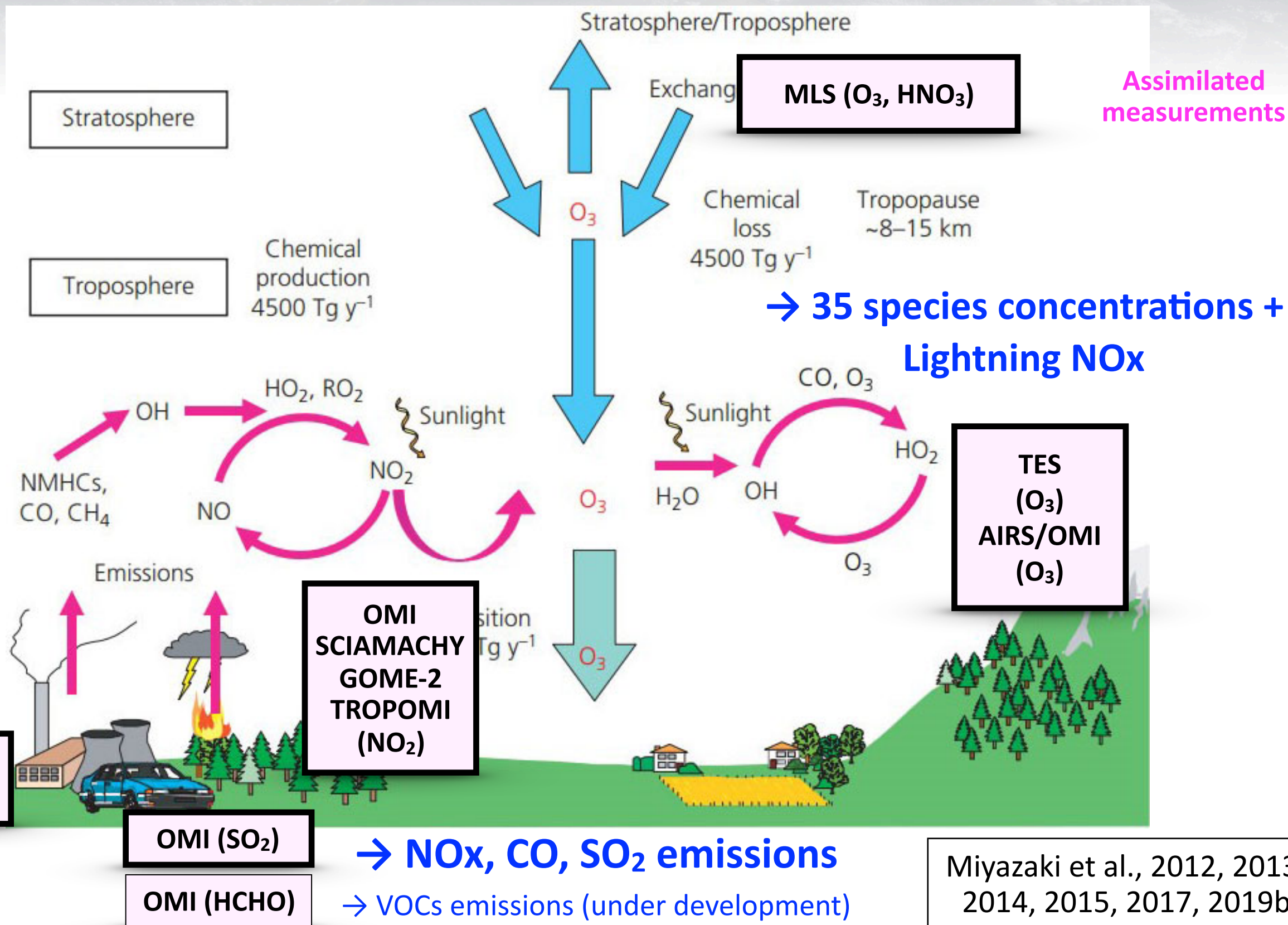
- **The in-situ observing network** is clearly insufficient for global health impact assessment especially in developing countries that suffer from severe air pollution.
- **Satellite measurements** have provided an unparalleled source of global data but suffer from limited surface sensitivity for many key species.
- These limitations can be mitigated by **new state-of-the-art chemical data assimilation systems** that optimize the information available in these datasets.



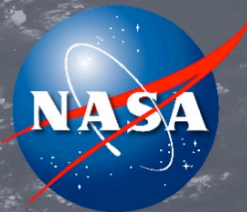


# Multi-constituent chemical data assimilation

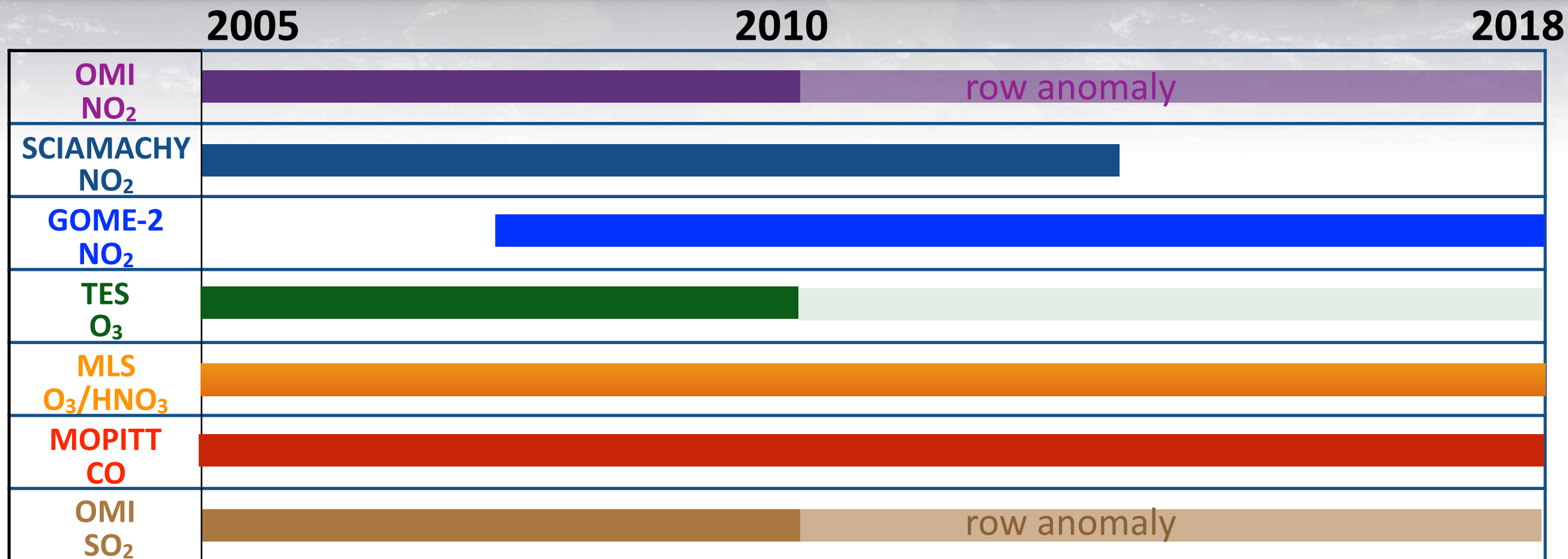
*EnKF data assimilation to integrate a suite of measurements from multiple satellite sensors*







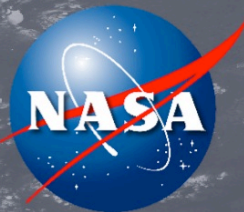
# Tropospheric chemistry reanalysis (TCR-2)



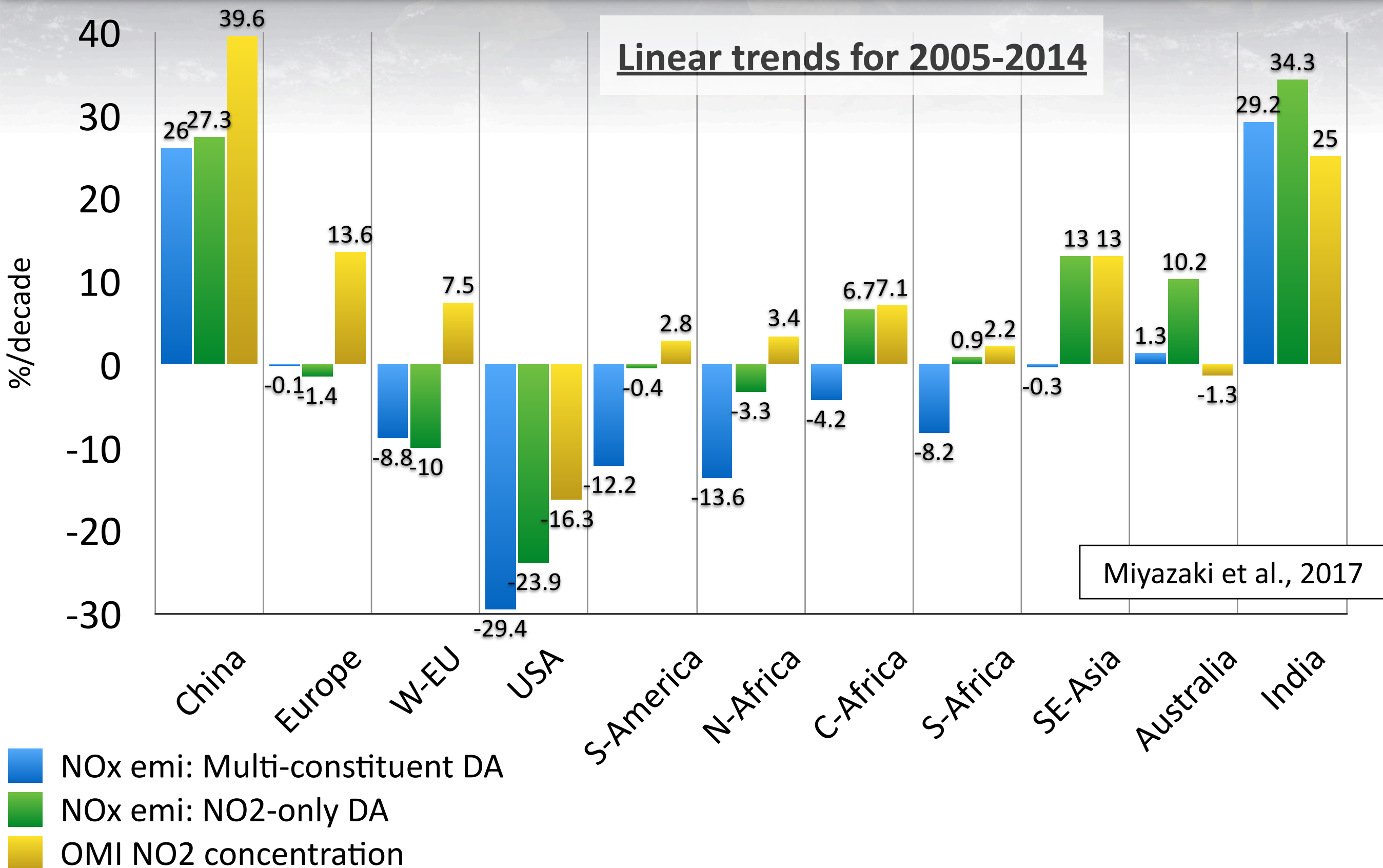
**Two-hourly,  
1.1° resolution,  
Emissions &  
concentrations**

- (1) understand the processes controlling the atmospheric environment
- (2) provide initial/boundary conditions for climate/chemical simulations
- (3) evaluate climate models and bottom-up emission inventories
- (4) suggest developments of models/observations (e.g., satellite concepts)

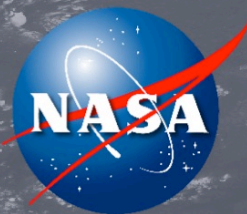




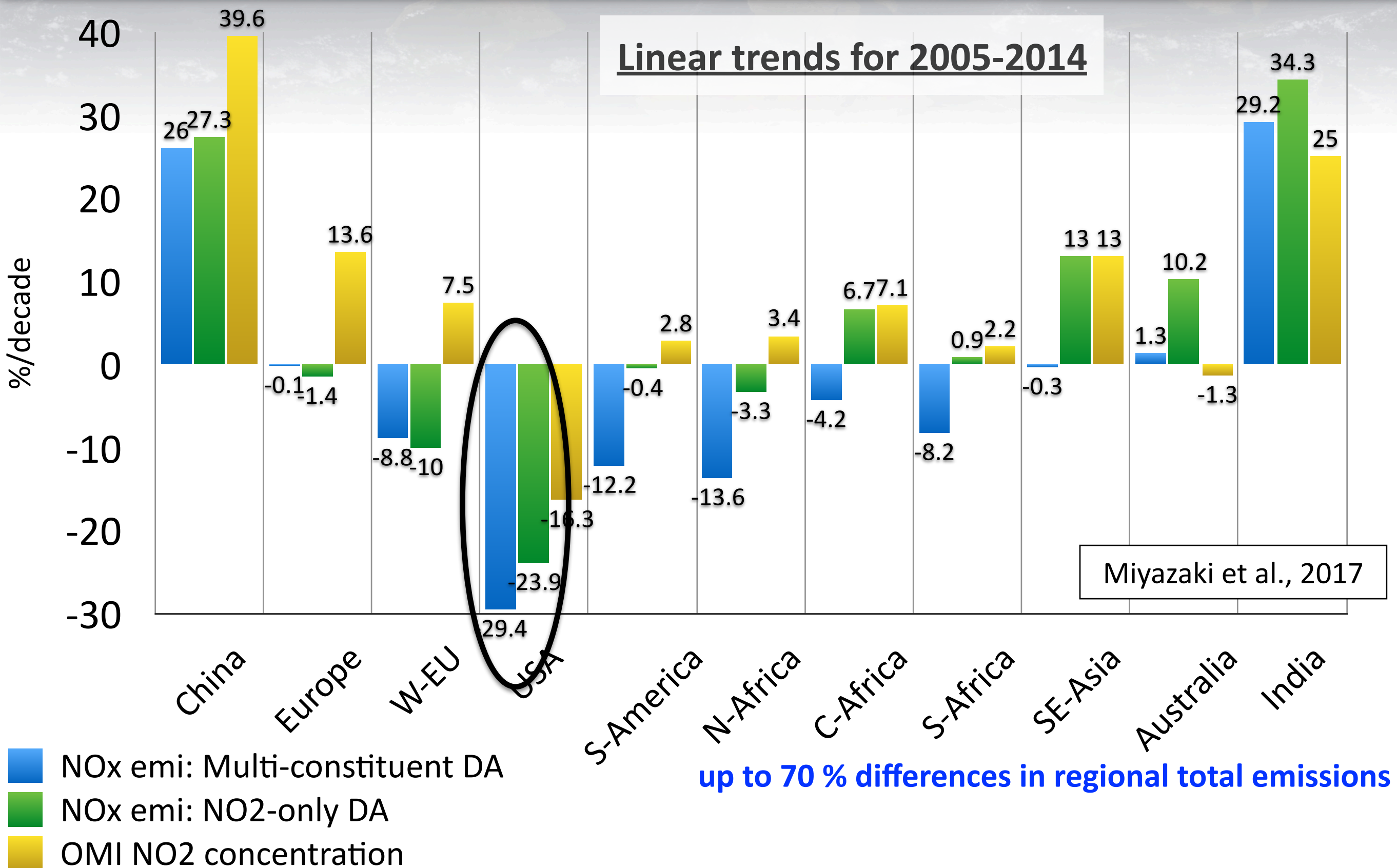
# Multi-constituent constraints on NO<sub>x</sub> emissions

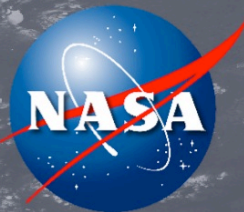




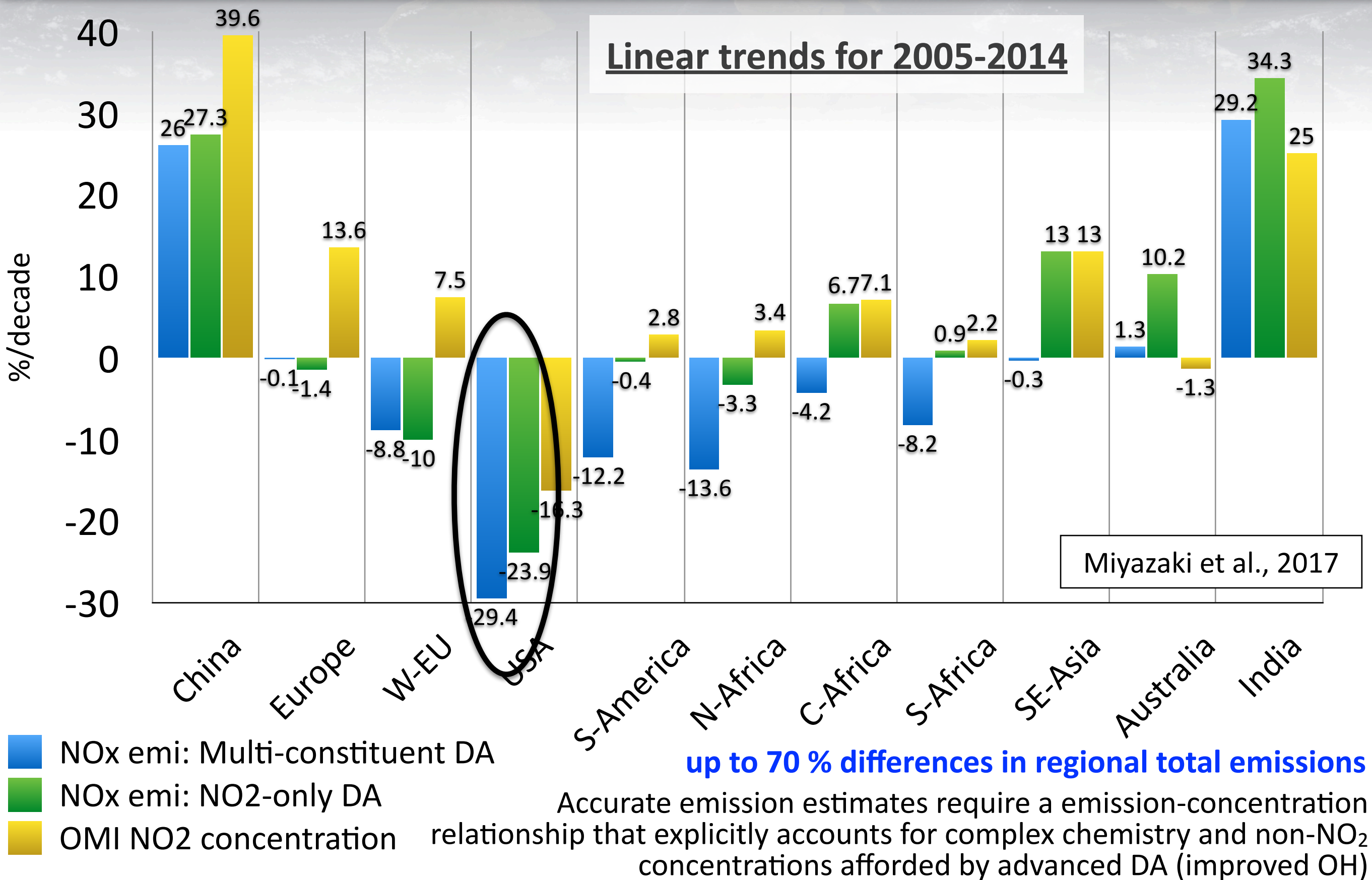


# Multi-constituent constraints on NO<sub>x</sub> emissions

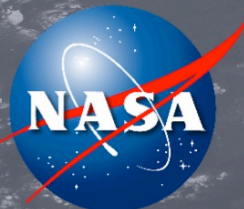




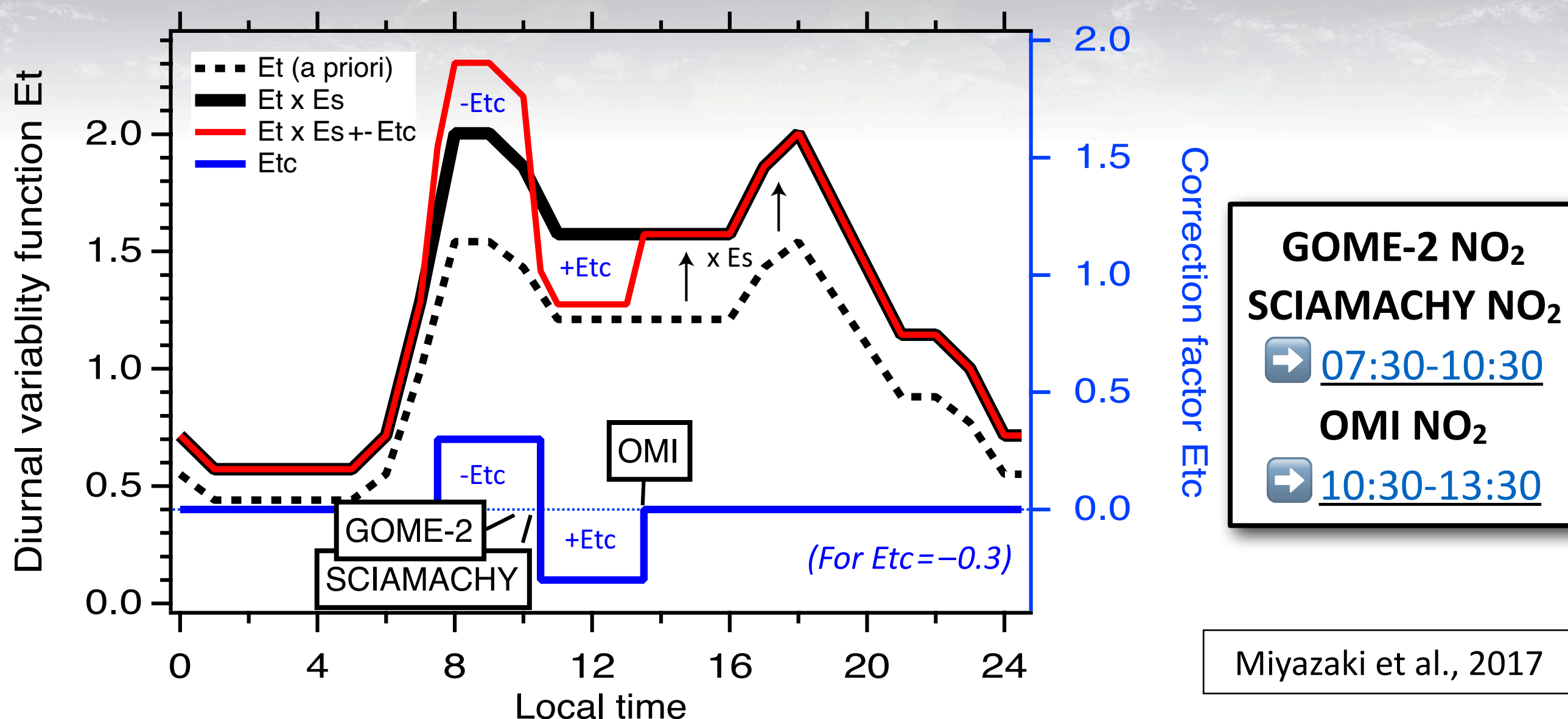
# Multi-constituent constraints on NO<sub>x</sub> emissions





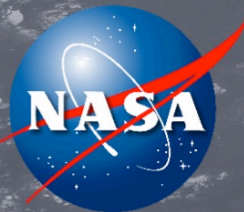


# Multi-sensor constraints on diurnal NO<sub>x</sub> emissions

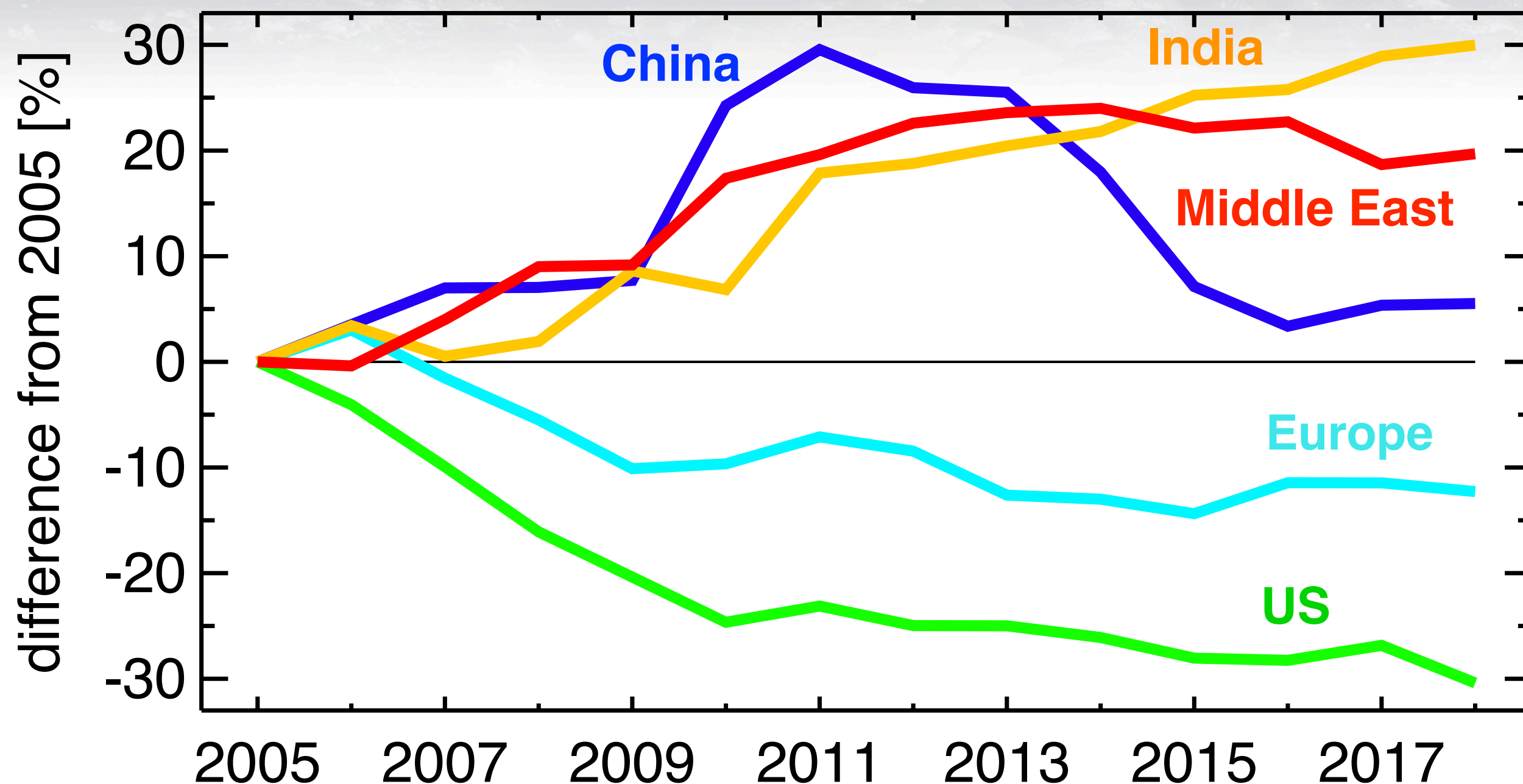


- A correction scheme is applied to modify the shape of the diurnal emission variability using multiple NO<sub>2</sub> measurements obtained at different overpass time.
- **Etc is mostly negative** -> A larger negative bias in simulated NO<sub>2</sub> in the morning. Larger underestimations in emissions (e.g., morning traffic rush) and/or larger model errors in chemical lifetime.



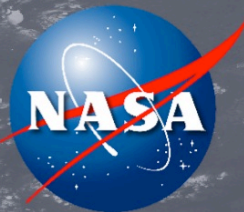


# Global NO<sub>x</sub> emission trends (2005-2018)

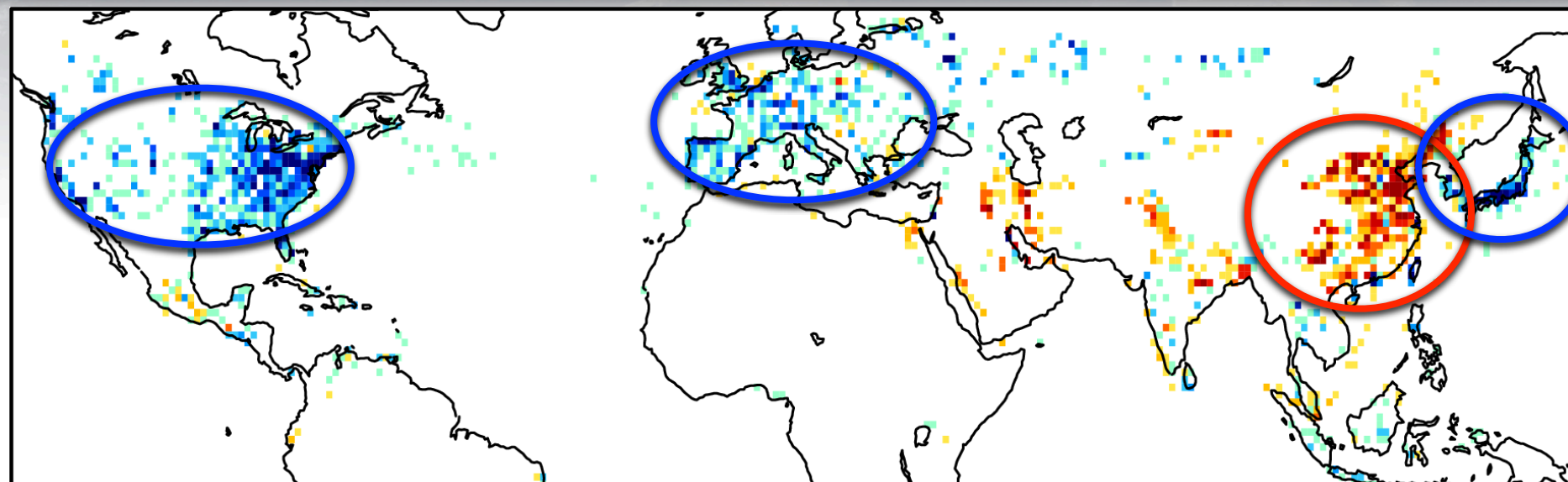


**Global total emissions:**

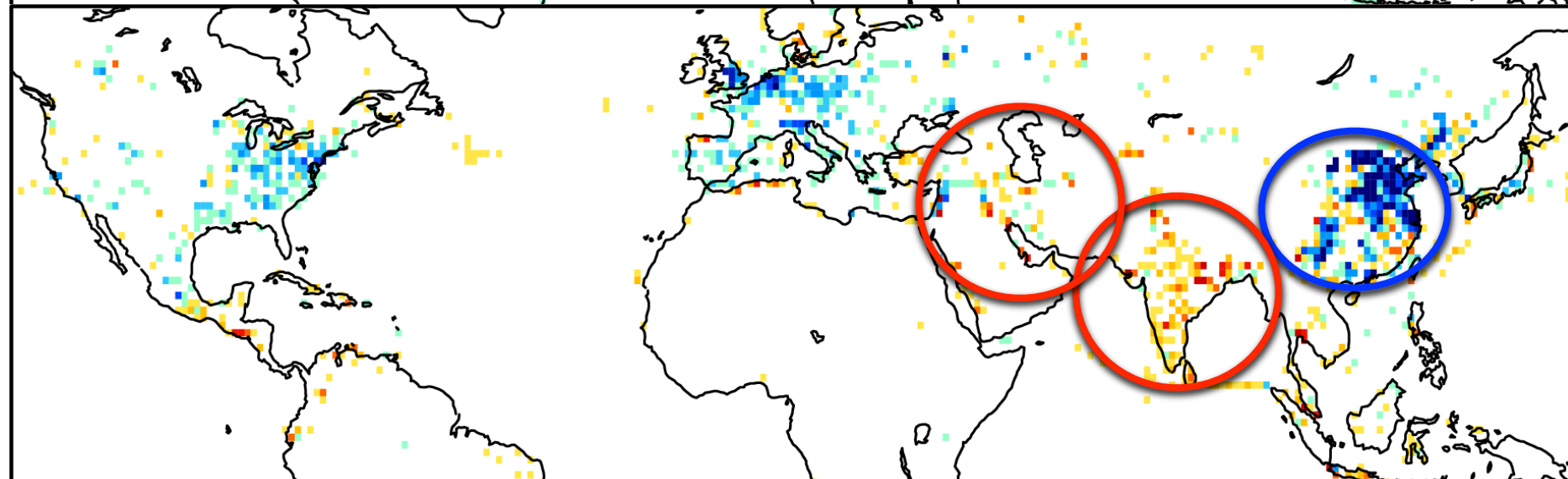
*Almost constant during 2005-2018 ( $49.3 \pm 2.7 \text{TgN}$ )*



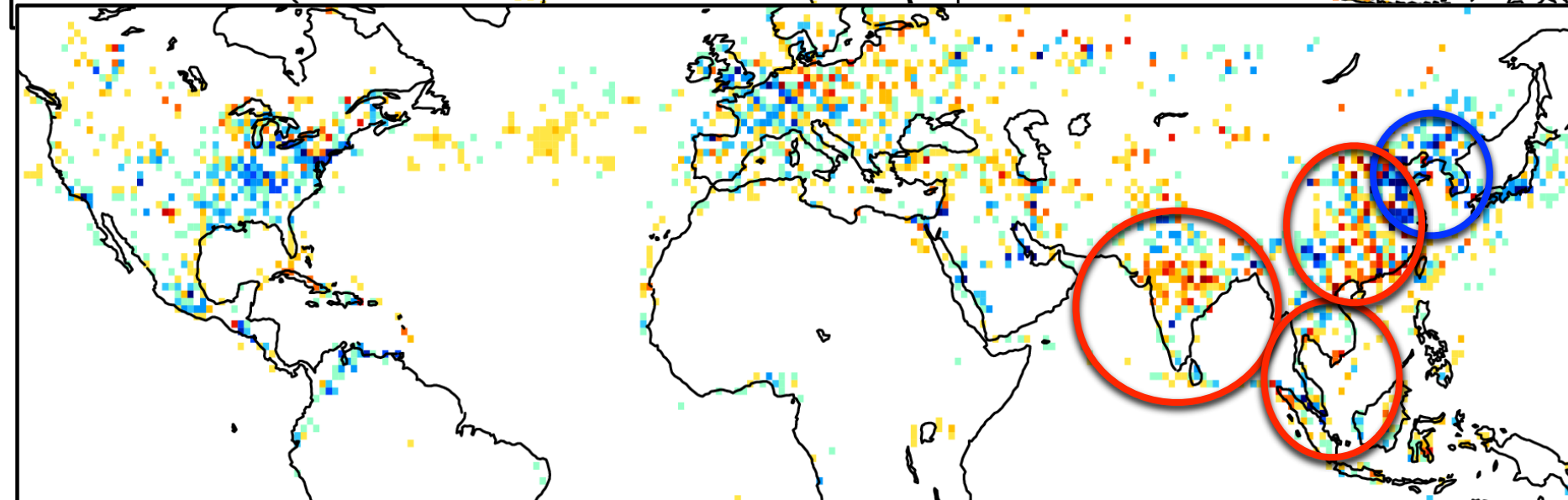
# Global NO<sub>x</sub> emission trends (2005-2018)



**2005-2010**



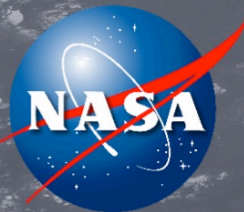
**2010-2015**



**2015-2018**

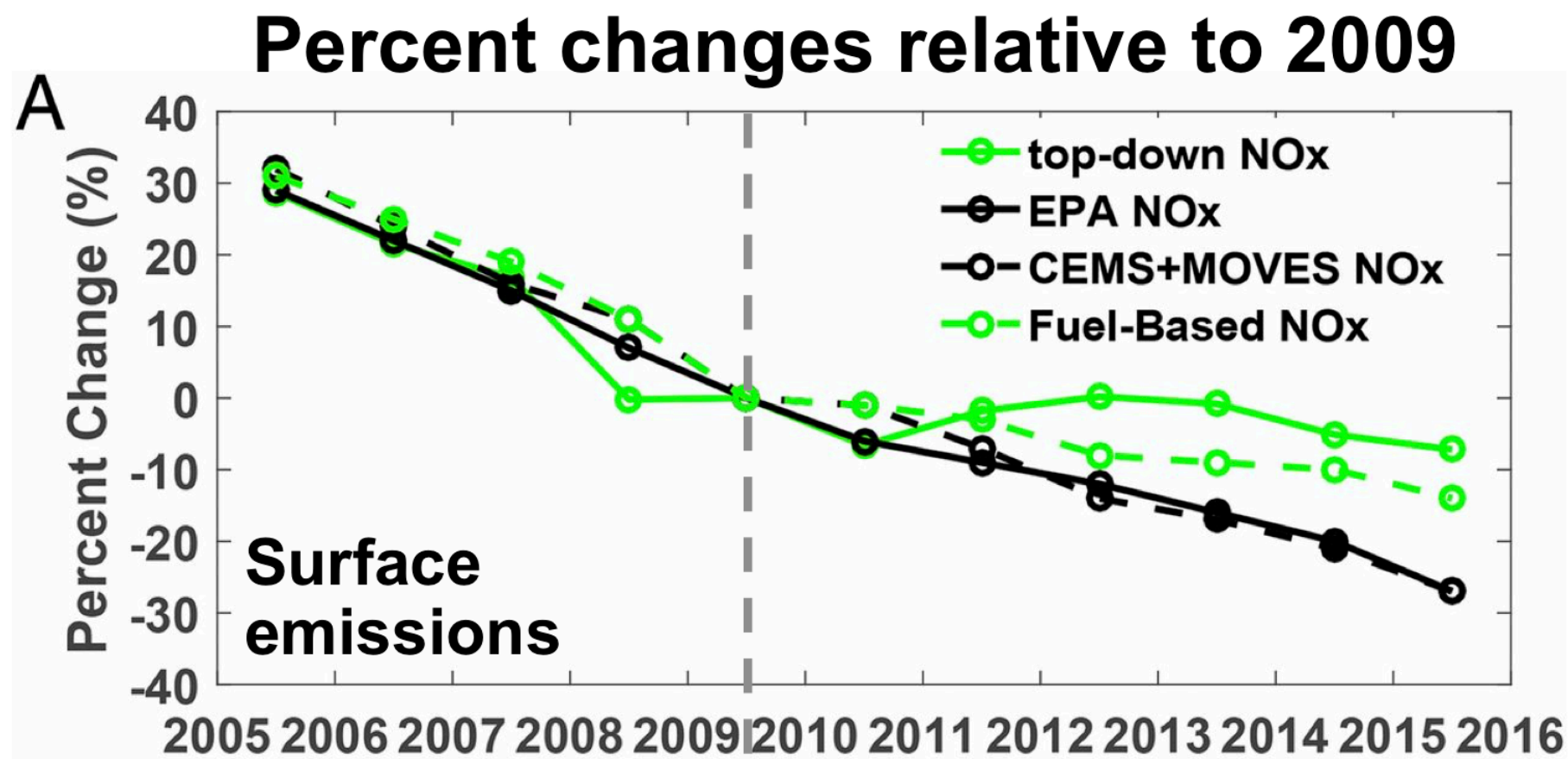
*strong impacts on air  
quality and human health  
in developing countries*





# Unexpected slowdown of US NO<sub>x</sub> emission reduction

Jiang et al.,  
PNAS, 2018



NO <sub>x</sub> top down	$-7.0 \pm 1.4\% \text{ a}^{-1}$	$-1.7 \pm 1.4\% \text{ a}^{-1}$
EPA NO <sub>x</sub>	$-6.4\% \text{ a}^{-1}$	$-5.3\% \text{ a}^{-1}$
Fuel based	$-6.7\% \text{ a}^{-1}$	$-2.9\% \text{ a}^{-1}$

*Fuel-based NO<sub>x</sub> bottom up estimates show some flattening in the trend.*

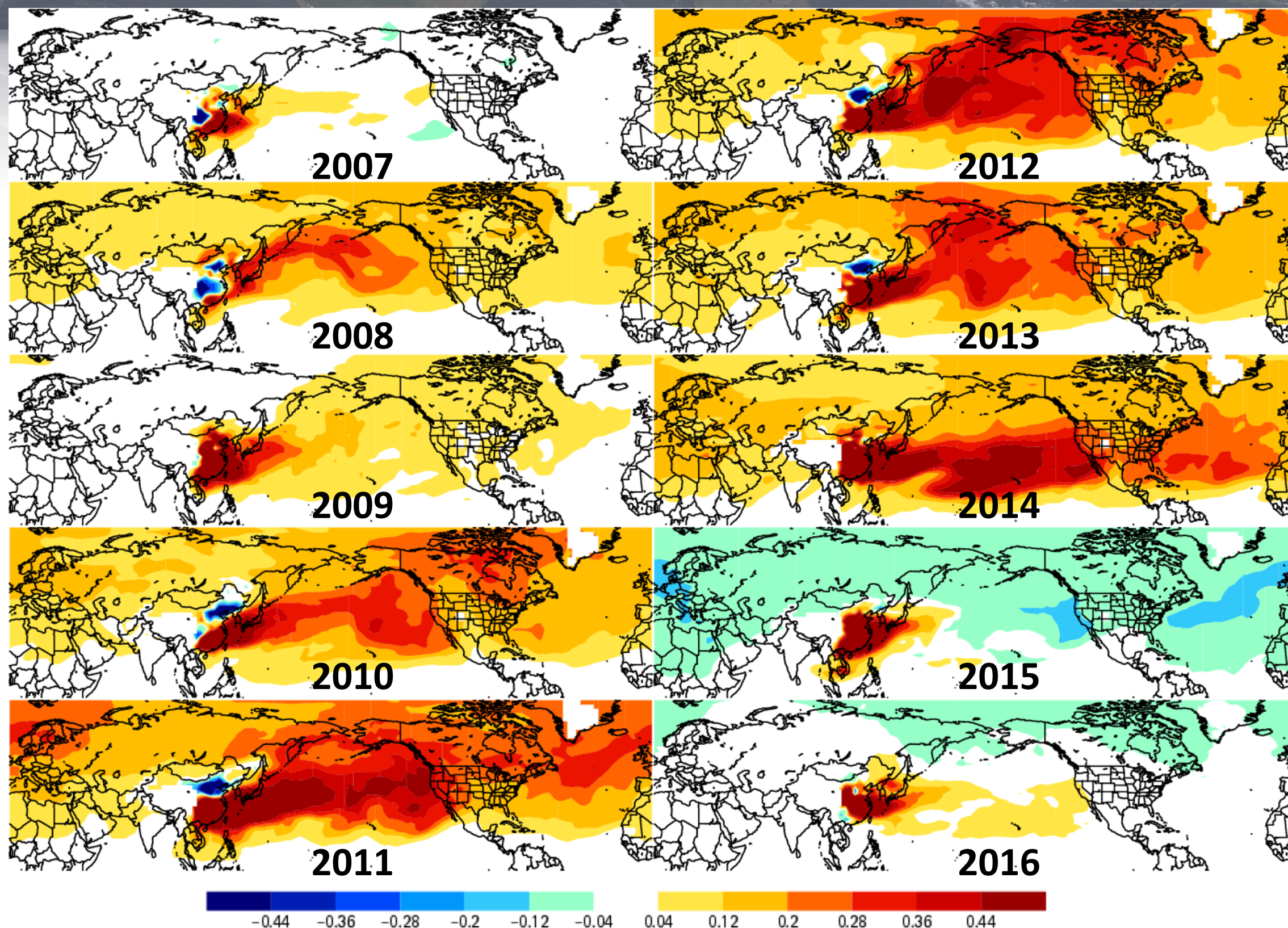
## Main contributions to fuel-based and NEI trend differences:

1. Off-road vehicles and area sources (industrial & residential) ↑
2. On-road diesel emissions not decreasing as expected ↑
3. On-road gasoline vehicles contributing fractionally less and maybe reaching diminishing returns ↓



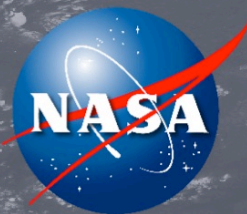


# HAQAST: US background ozone



**750 hPa mean ozone anomaly in April due to Chinese NO<sub>x</sub> emission changes from 2006 demonstrate the importance of local/remote emissions on air pollution exposure assessment**

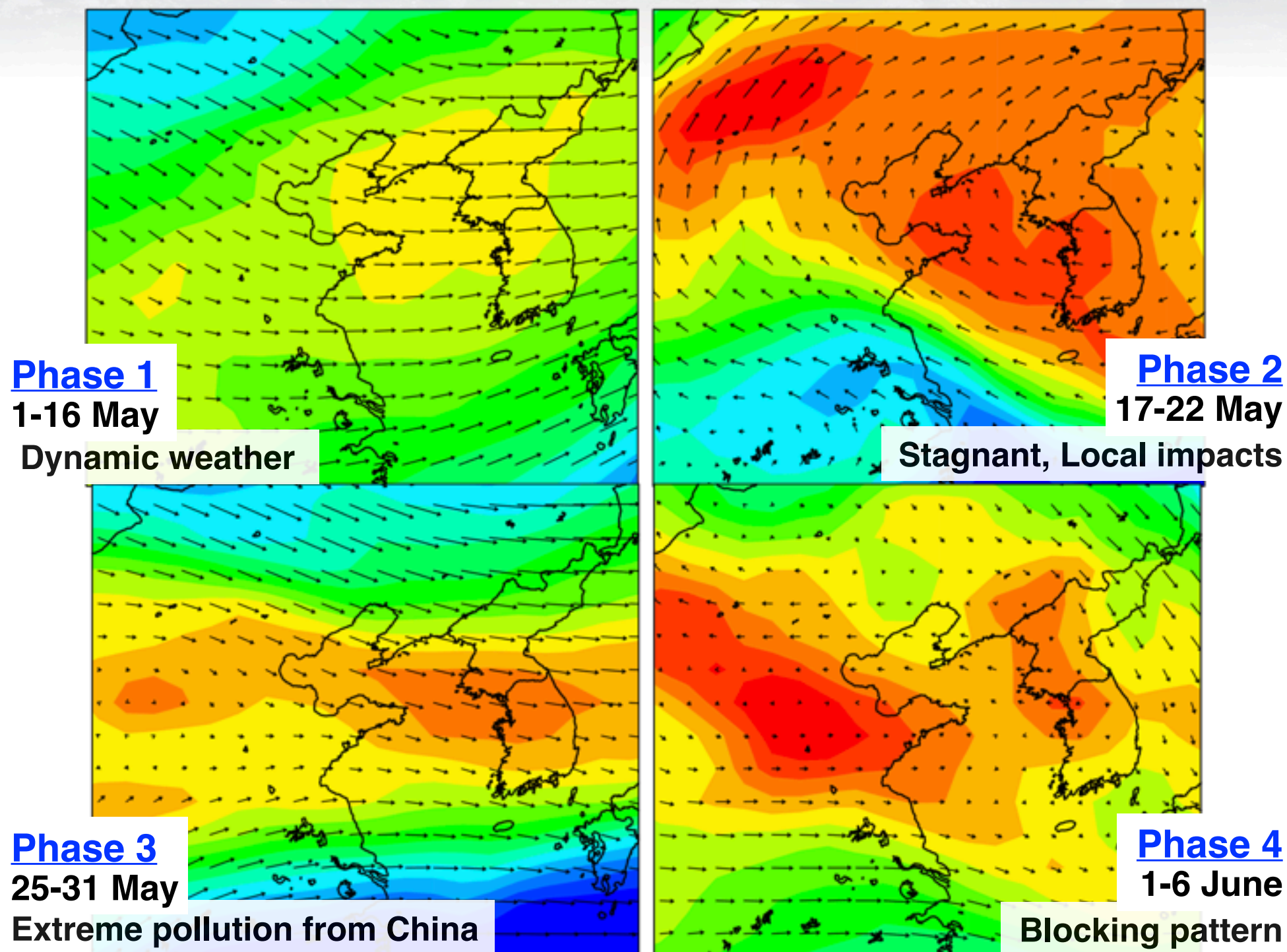




# KORUS-AQ aircraft campaign (May 2016)

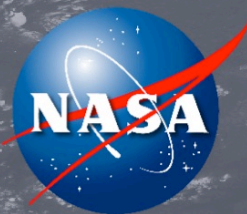


## Reanalysis ozone at 700 hPa



*Dynamic changes in air pollution*





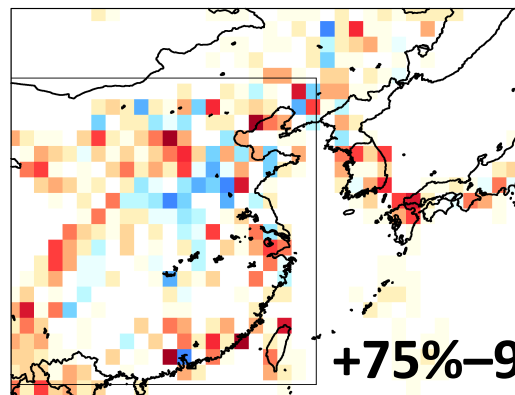
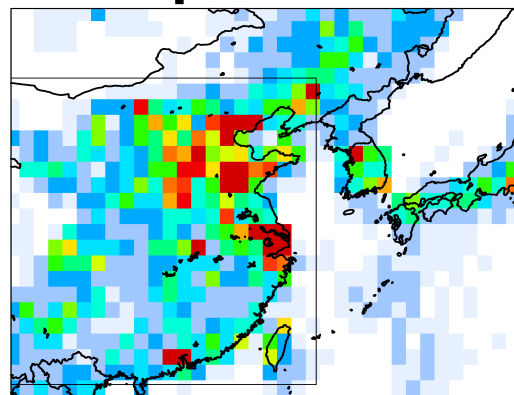
# KORUS-AQ aircraft campaign (May 2016)



## Emission estimates

### A posteriori

### Increments

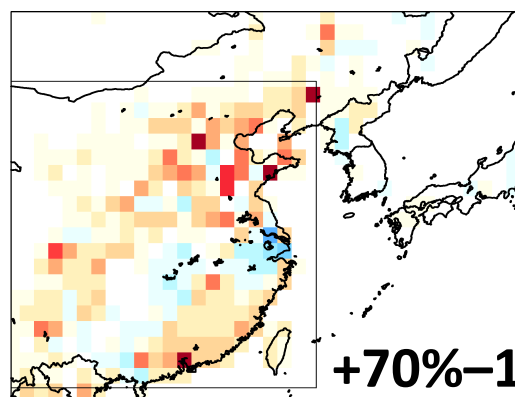
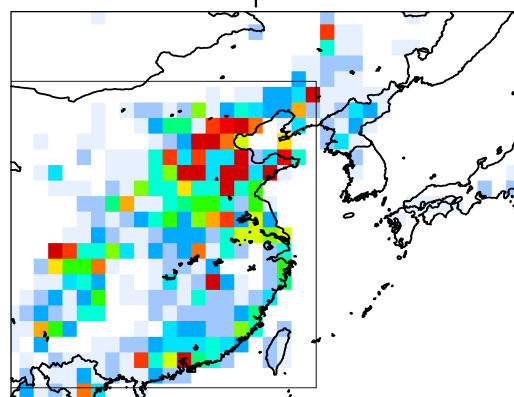


+75%–94%

NO<sub>x</sub>

### CO : A posteriori

### CO : Increment

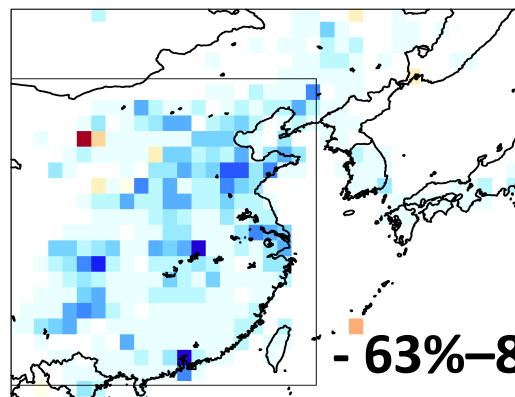
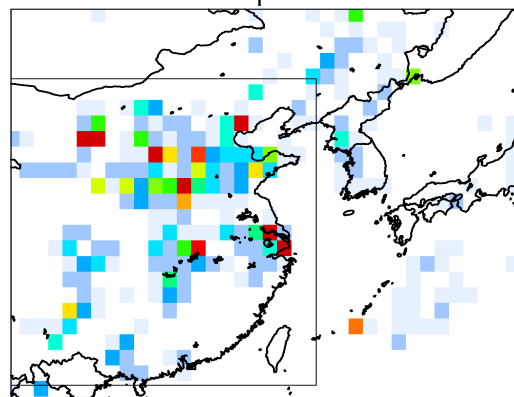


+70%–140%

CO

### SO<sub>2</sub> : A posteriori

### SO<sub>2</sub> : Increment



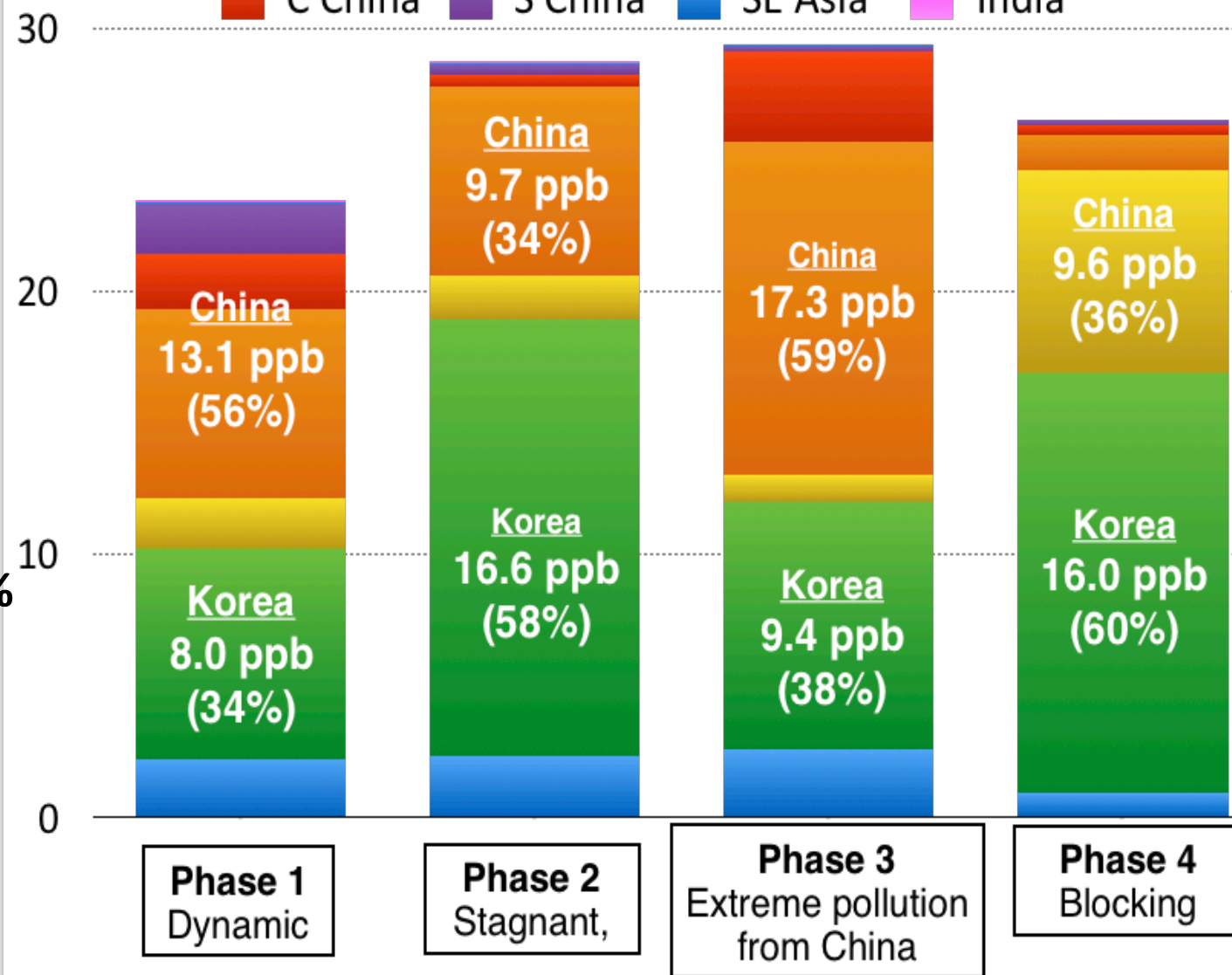
- 63%–83%

SO<sub>2</sub>

Miyazaki et al., 2019a

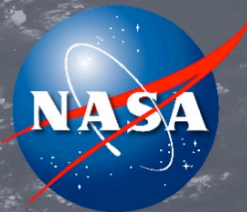
## Source-receptor analysis

*from NO<sub>x</sub> emissions to O<sub>3</sub> over Seoul at 900 hPa*



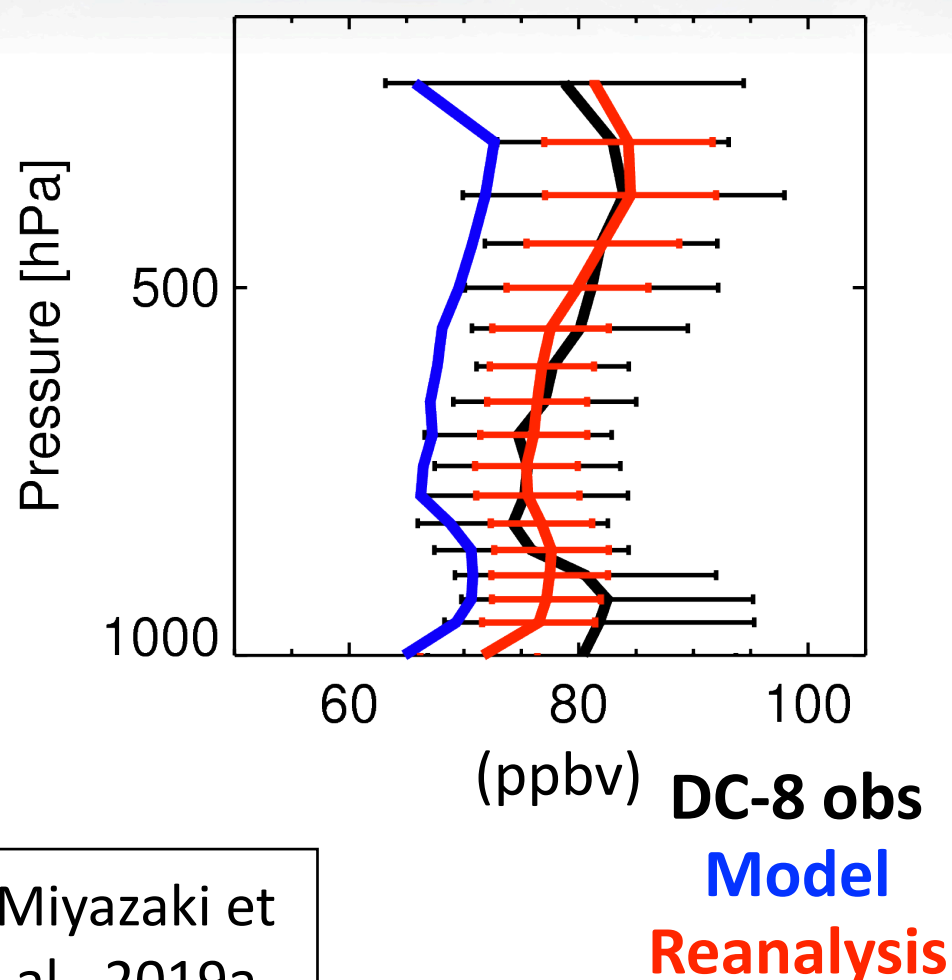
**Dynamic changes in emission influences  
for effective ozone control strategy (+VOCs)**





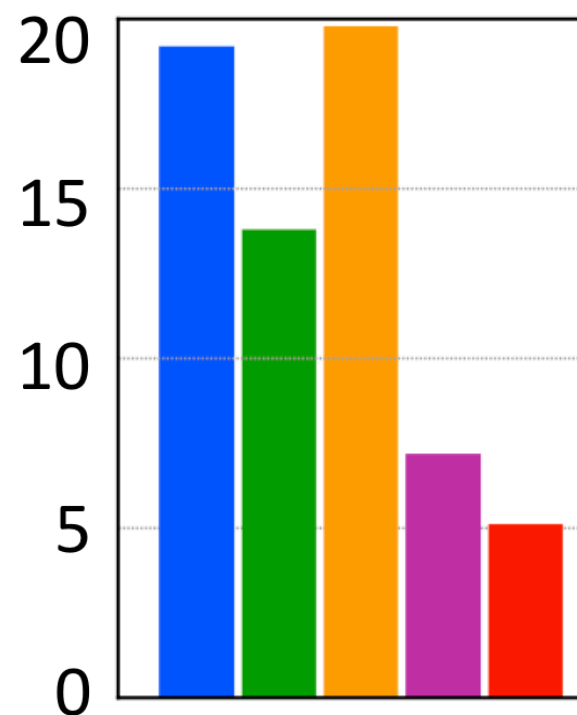
# Impact of individual assimilated measurements

## Mean ozone profile



Miyazaki et al., 2019a

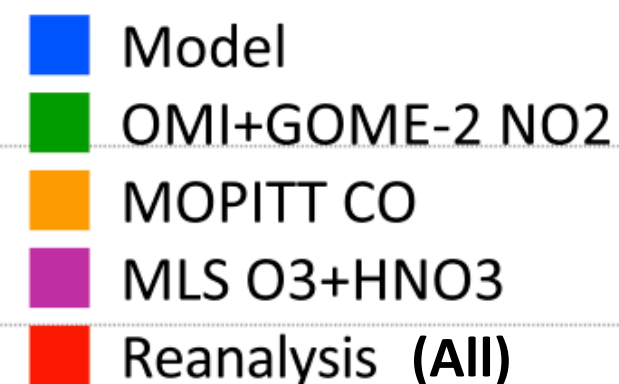
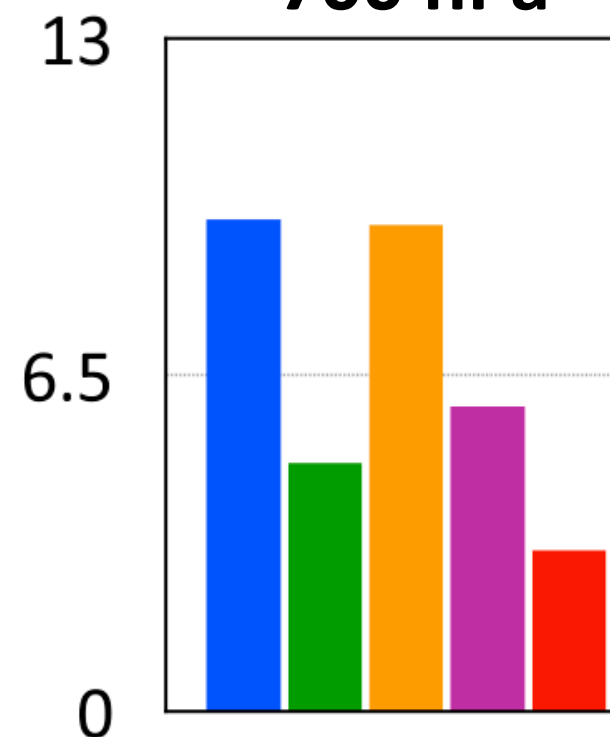
## 400 hPa



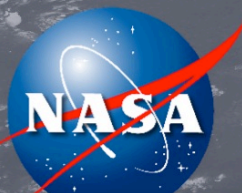
**Ozone biases to DC-8 (ppb)**  
(during stagnant condition)

*Observing System  
Experiments (OSEs)*

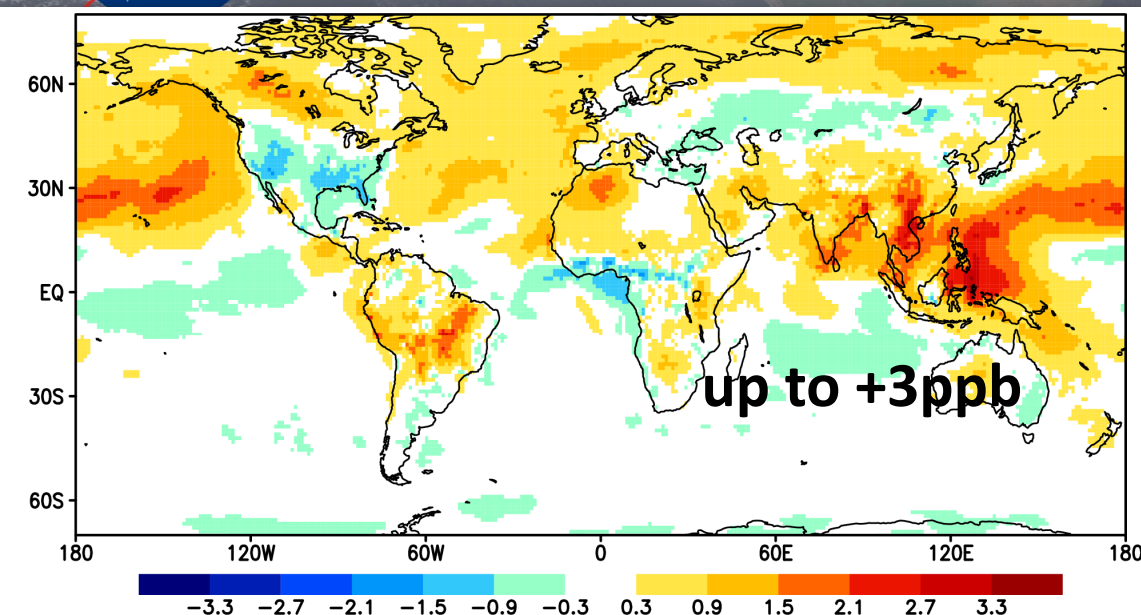
## 700 hPa



- **OMI + GOME-2 NO<sub>2</sub>** → Improved the lower tropospheric ozone
- **MLS O<sub>3</sub>/HNO<sub>3</sub>** → Additional important corrections throughout the troposphere.
- **Multi-constituent (Reanalysis)** → correct the entire tropospheric ozone profile
- **AIRS/OMI ozone** (not shown) → further improvements for any met condition.



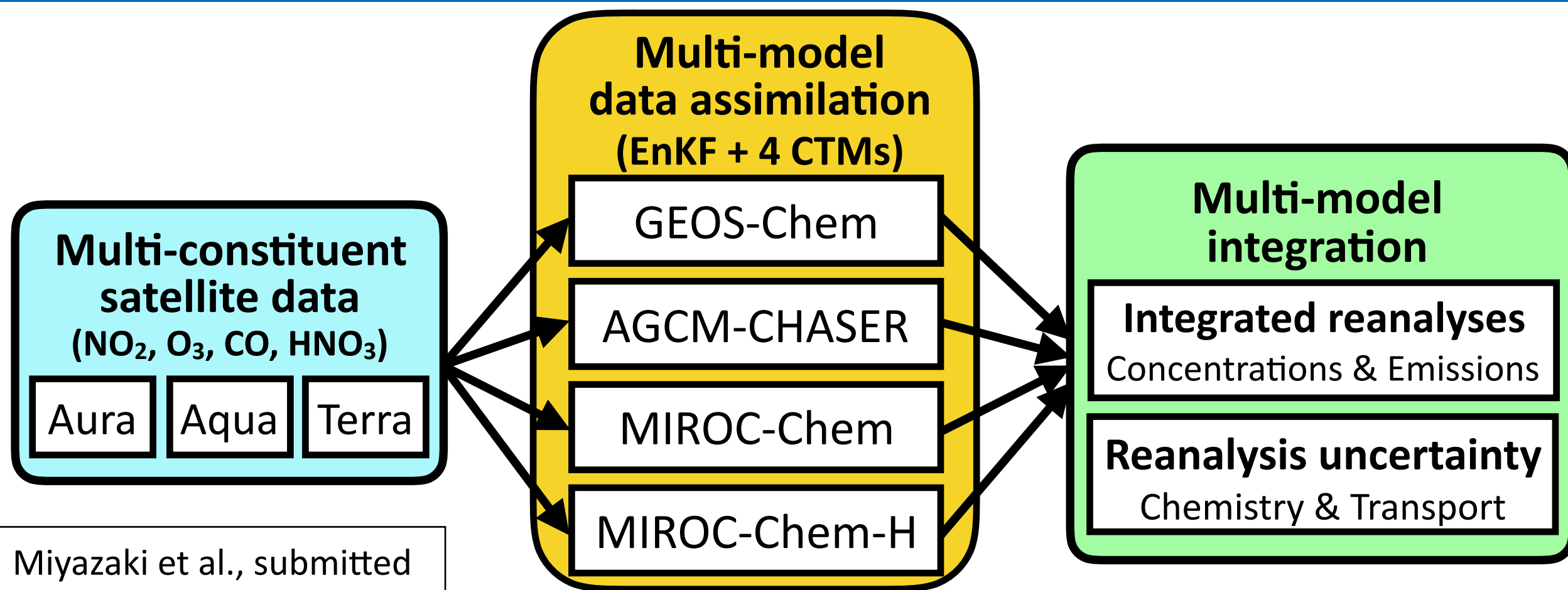
# Multi-model data assimilation integration



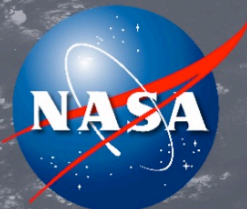
**Surface ozone changes [ppbv] due to NO<sub>x</sub> emission changes: 2010-2016 minus 2005-2009**

Chemical reanalyses provide useful information on exposure estimates and its attributions. Nevertheless, **systematic model errors must be quantified in order to assess their fidelity for exposure studies.**

## Multi-mOdel Multi-cOnstituent CHEMical data assimilation (MOMO-Chem)







# Multi-model data assimilation integration

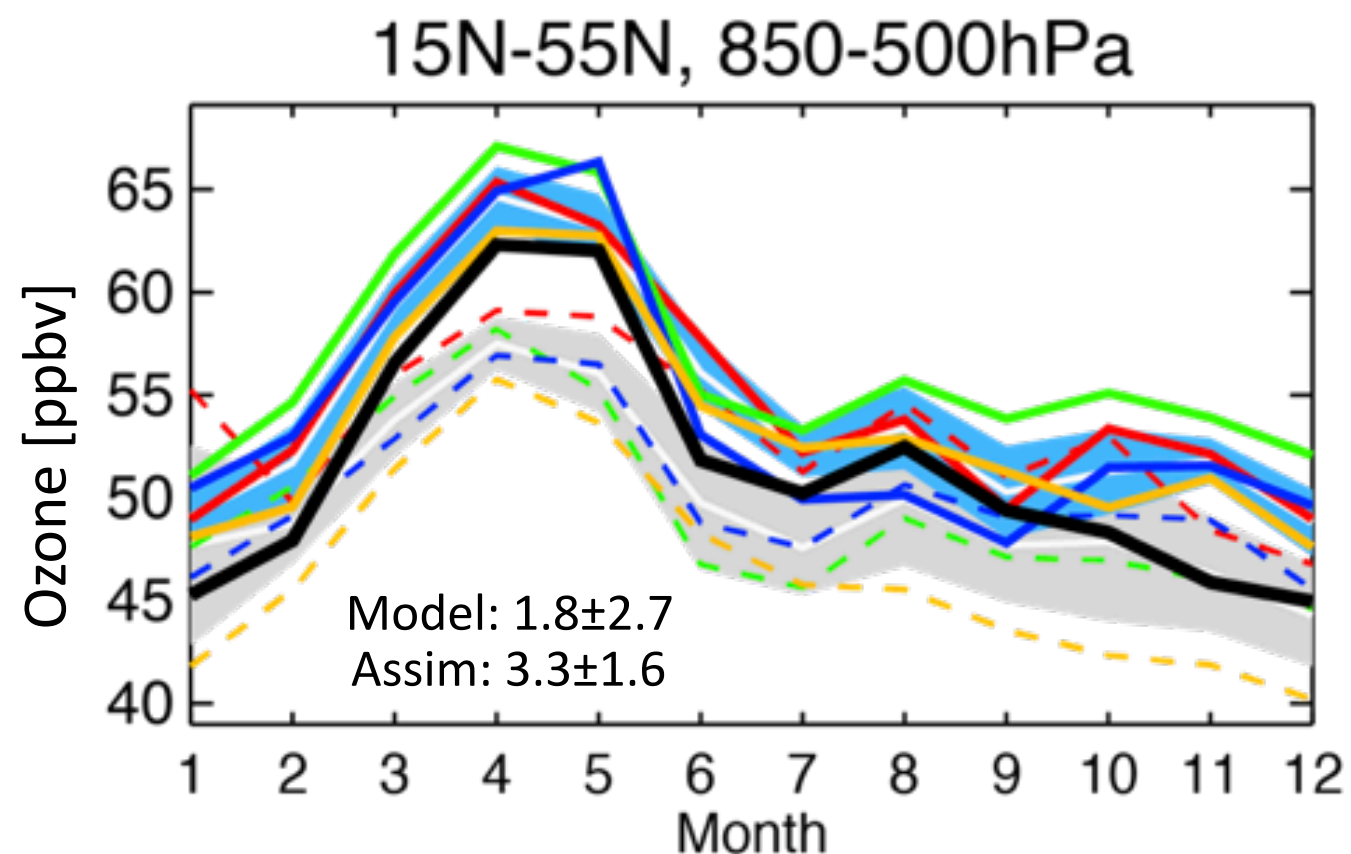
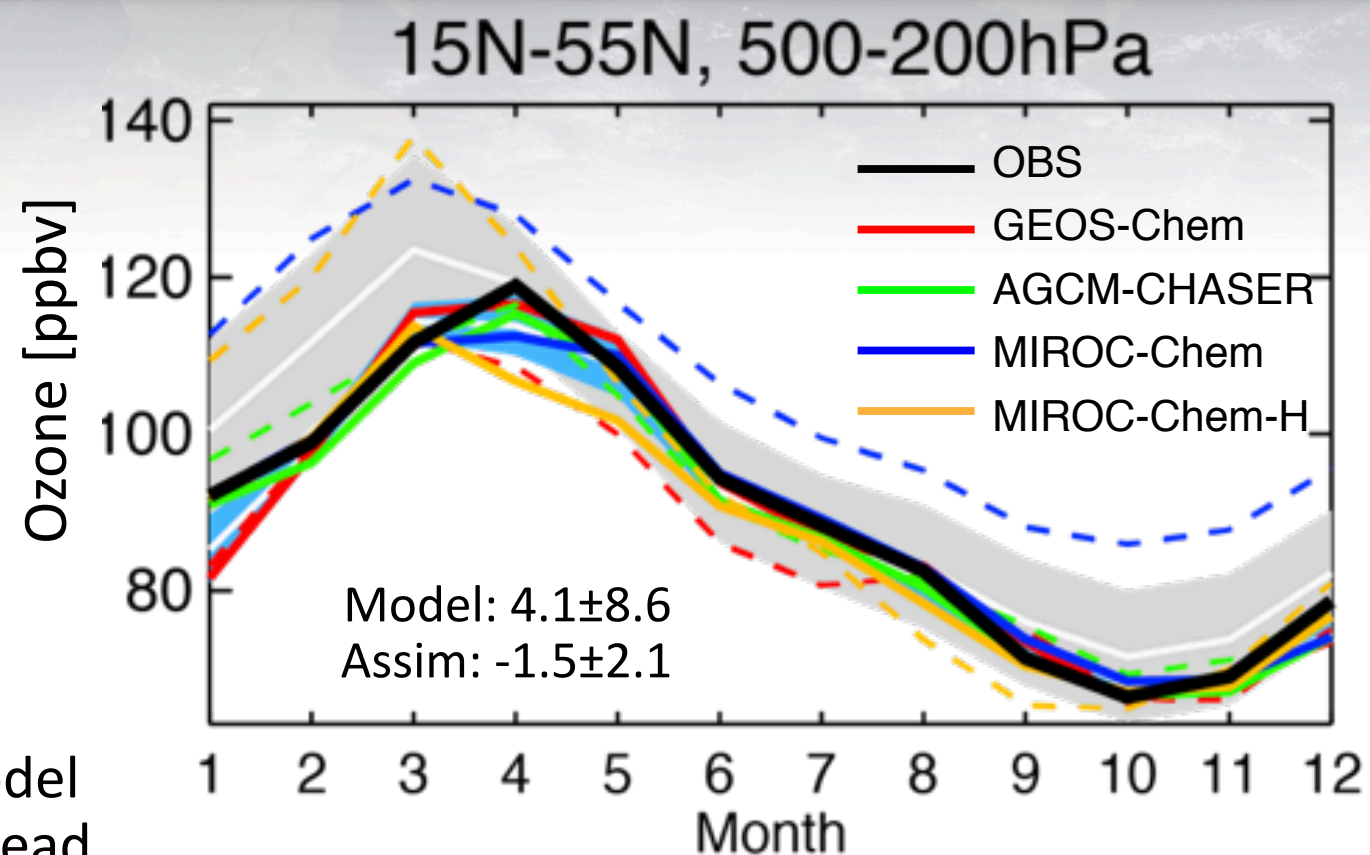
**Multi-constituent assimilation greatly reduced the annual mean model bias (by 40-80% in NH, 50-90% in TR, 45-95% in SH) and the multi-model spread (by 20-60% in NH and 30-85% in SH)**

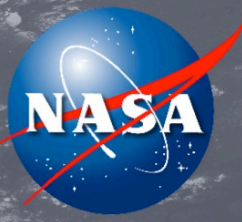
— Assimilation  
..... Model  
— Multi-model mean/spread

Harnessing the current observing system provides sufficient constraints to greatly reduce the influences of model errors and to provide the consistent concentration analysis for O<sub>3</sub>, NO<sub>2</sub>, CO, and OH.

Miyazaki et al., submitted

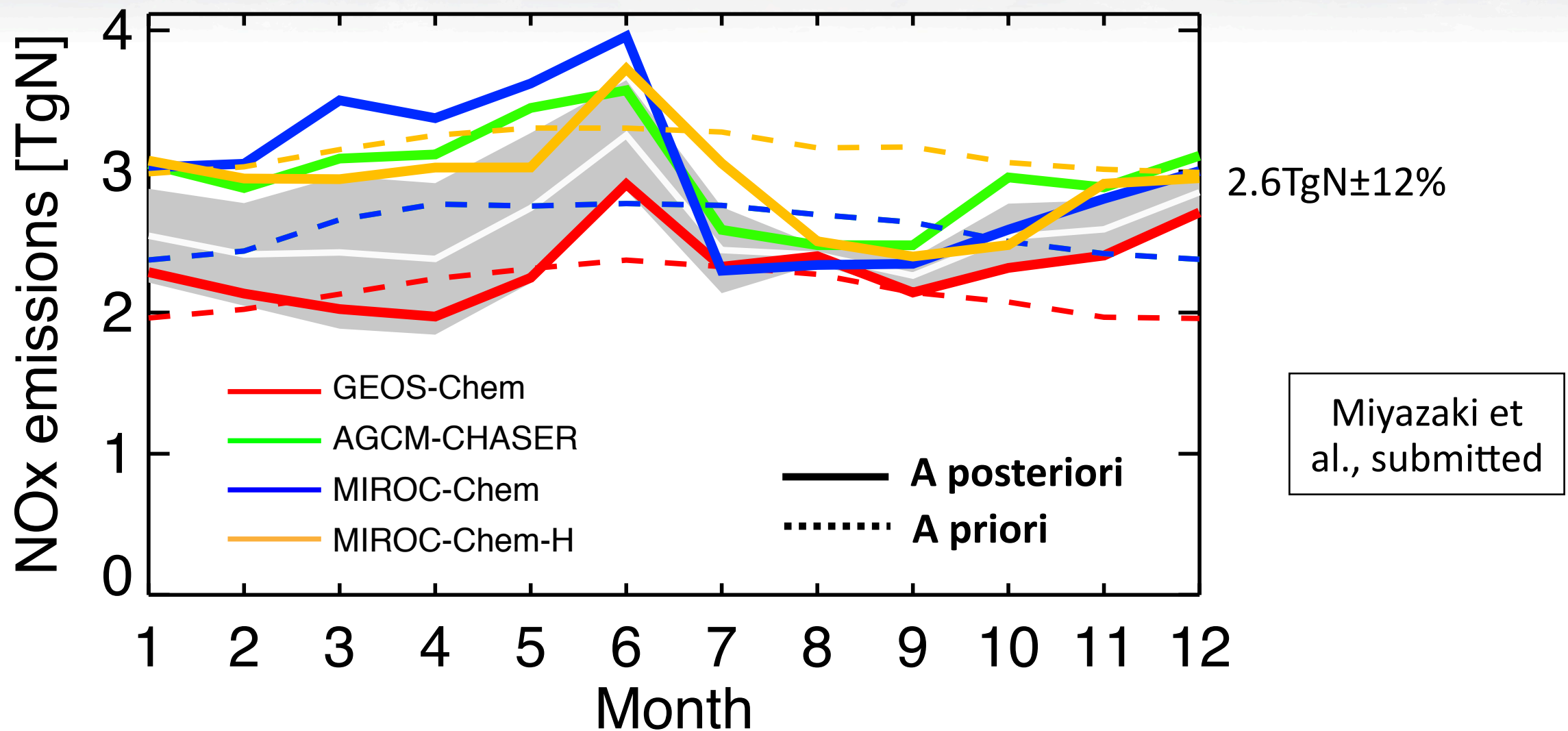
vs ozonesonde





# Multi-model data assimilation integration

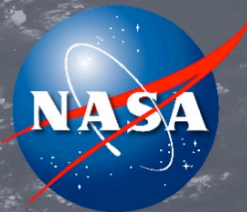
## NO<sub>x</sub> emissions : India



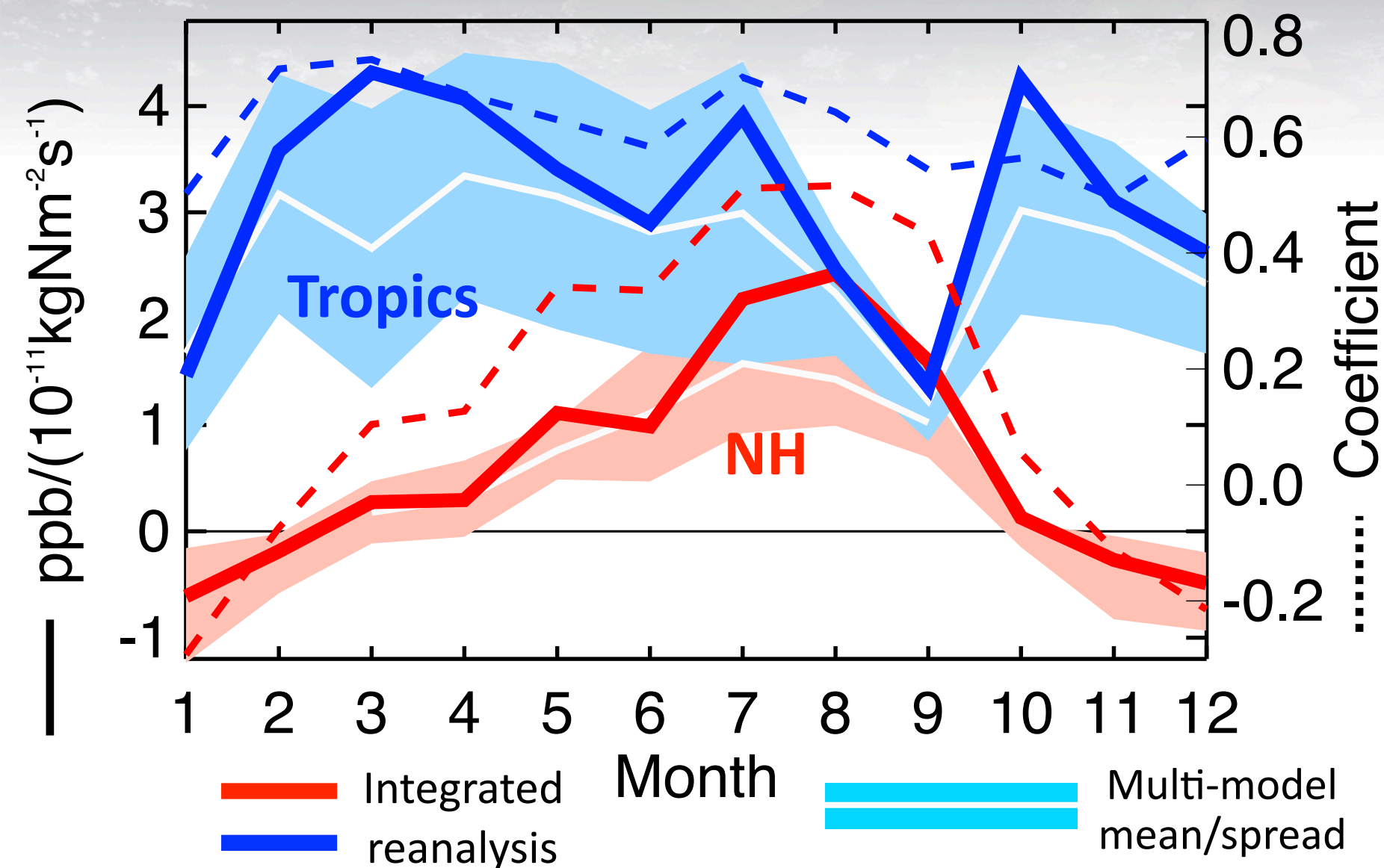
- Commonly suggests potential problems in the bottom-up emission inventories
- Possible uncertainty ranges in the a posteriori emissions due to model errors: 13–31% for industrialized areas and 4–21% for BB areas

→ **Different model responses to emissions among the systems**





# Multi-model data assimilation integration

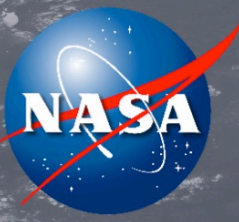


## Surface ozone response to NOx emissions

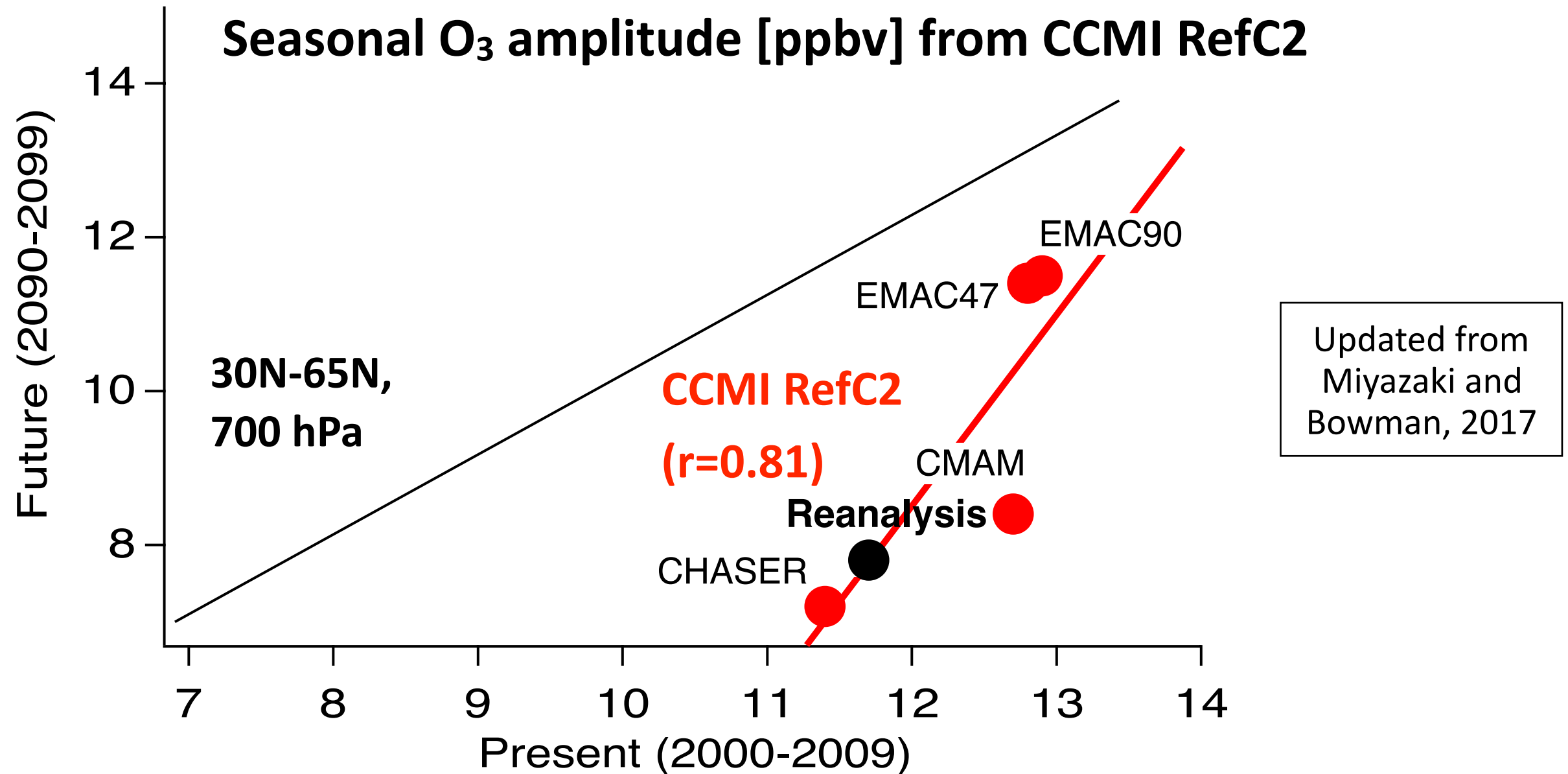
The analysis increments were used to quantify model sensitivities related to chemistry and transport

Miyazaki et al., submitted

- $\text{TR} > \text{NH}$  : Latitudinal shifts in NOx emissions would increase global ozone.
- The observationally-constrained, multi-model integrated fields provide fundamentally different fast chemical processes than those in the individual models.
- This would provide insights into ozone production processes to inform chemical predictions through relationships such as emergent constraints

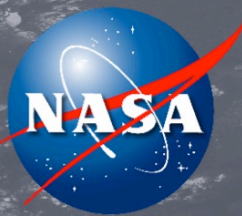


# For evaluation of chemistry predictions

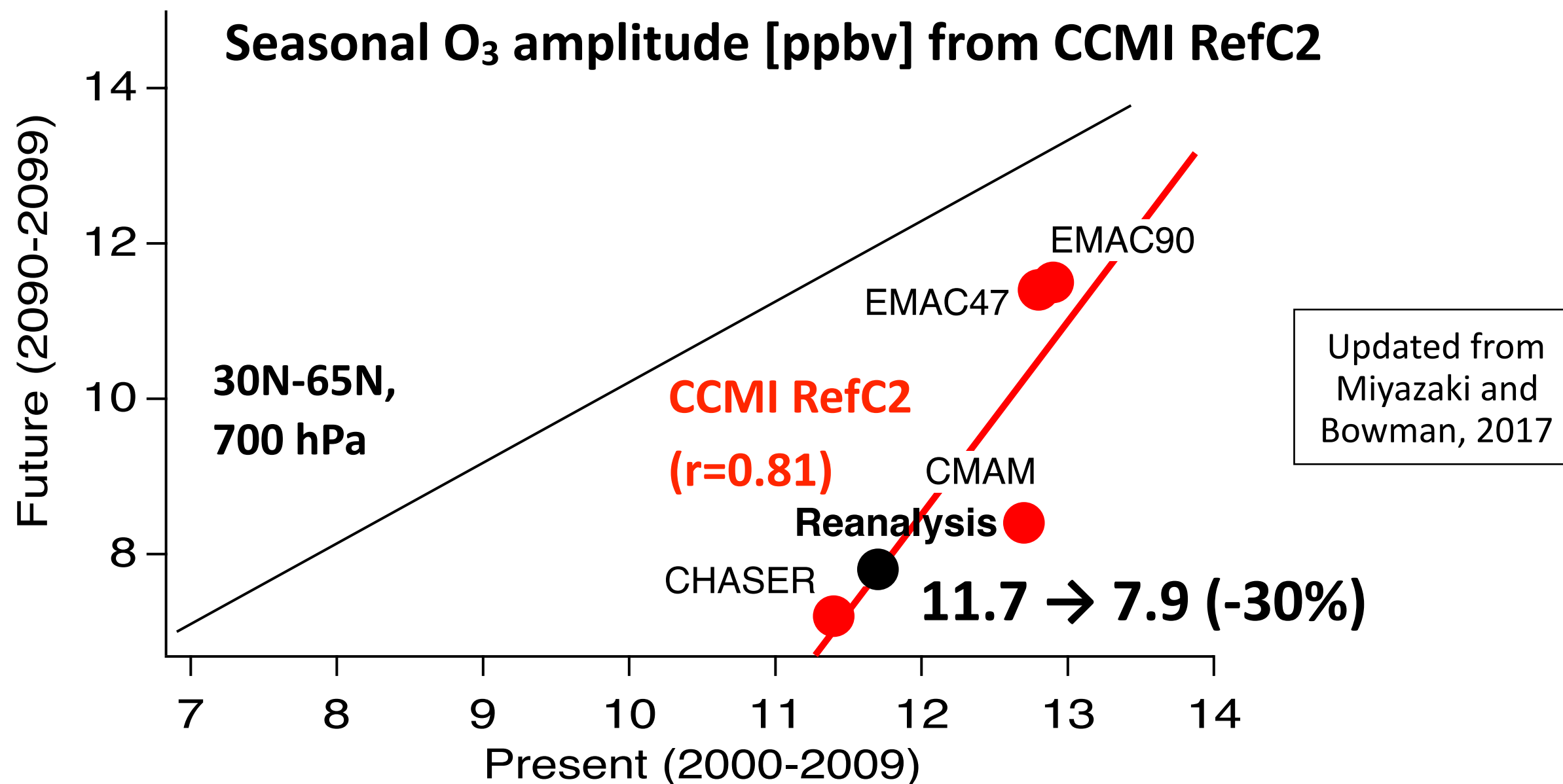


- To ensure more accurate future ozone/radiative forcing predictions by evaluating future simulations by using the reanalysis as emergent constraints.
- The similar approach could be applied to evaluate model responses to changing emissions in future climate predictions using the ozone response quantified in MOMOR-Chem.

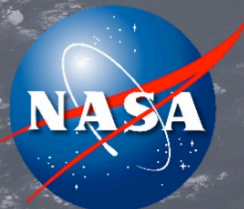




# For evaluation of chemistry predictions



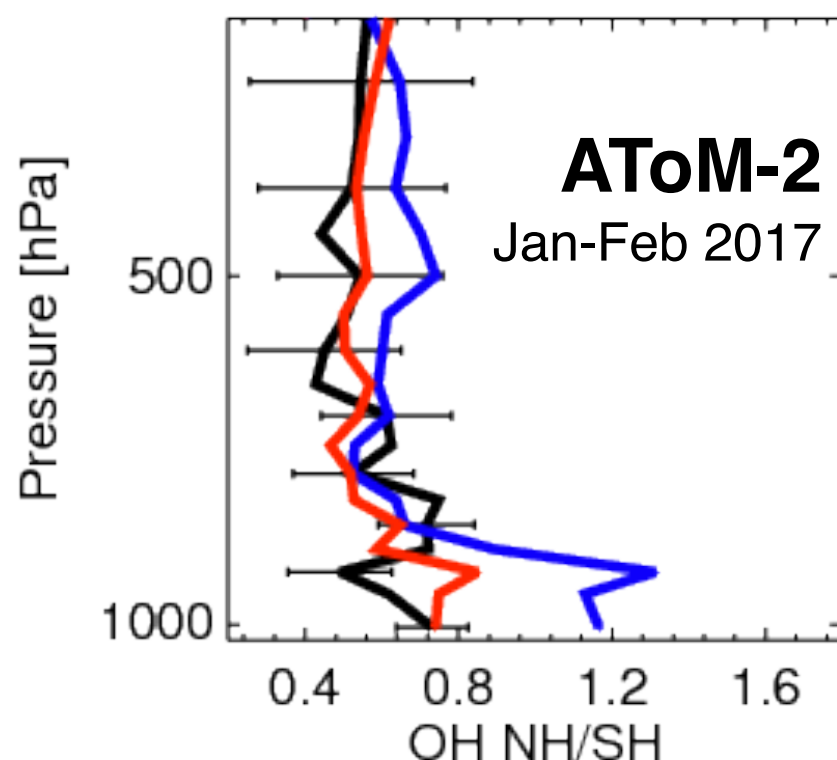
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# For evaluation of chemistry predictions

*Oxidation capacity response  
to changing emissions*

**OH NH/SH ratio**



MOMO-Chem mean $\pm$ stdev

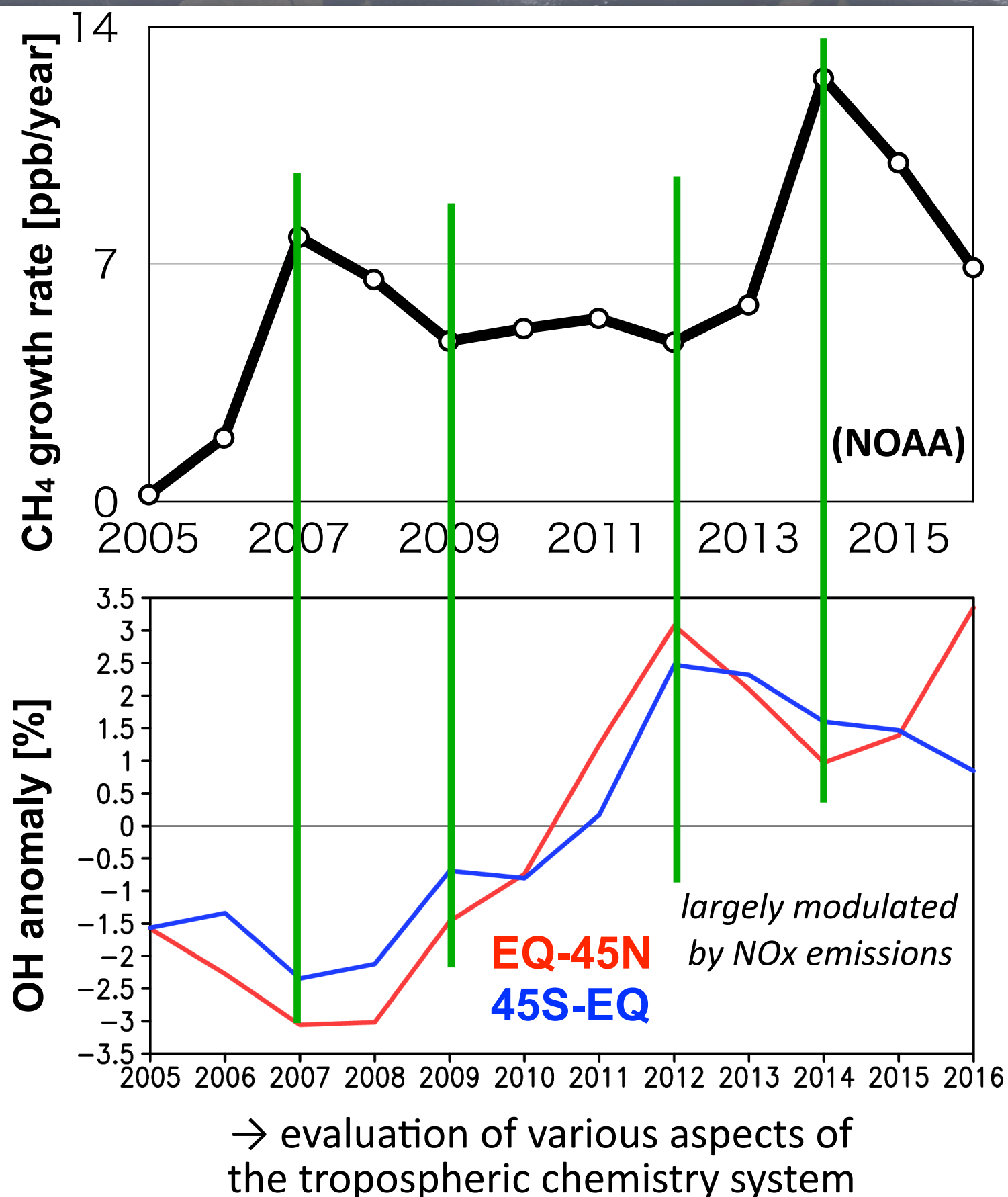
**Model**

**$1.29 \pm 0.03$**

**Assim**

**$1.18 \pm 0.03$**

Observational estimate = 0.97  
(Patra et al., 2014)





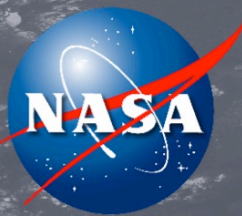
The reanalysis products will be distributed through the JPL TES website and GES DISC!  
*More than 20 variables, two-hourly, 14 years, 1.1 degree, 27 levels, global*

# TES

TROPOSPHERIC  
EMISSION SPECTROMETER

- **Chemical reanalysis products:** TCR-1 (Miyazaki et al., 2015), TCR-2 (Miyazaki, et al., in prep)
- **Air quality studies:** KOURUS-AQ (Miyazaki et al., 2019a; Thompson et al., 2019), Remote oceans (Kanaya et al., 2019), the Middle East ozone (Jiang et al., 2016), SE Asia ozone (Ogino et al., to be submitted), Machine learning (He et al., 2019)
- **Satellite data evaluations:** TES (Cady-Pereira et al., 2017), AIRS/OMI (Fu et al., 2018), IASI-GOME-2 (Cuesta et al, 2018), TES/OMI (in prep), QA4ECV OMI (Boersma et al., 2018)
- **Chemistry-climate model evaluations:** ACCMIP (Miyazaki and Bowman, 2017), CCMI (Kuai et al., 2019)
- **Emission inter-comparisons:** NO<sub>x</sub> (Ding et al., 2017; Miyazaki et al., 2014; 2017; 2019a, Itahashi et al., submitted; Elguindi et al., in prep), NO<sub>x</sub>/CO (Tang et al., 2019), Lightning NO<sub>x</sub> (Miyazaki et al., 2014)
- **Reanalysis comparisons:** CAMS ozone (Huijnen et al., submitted), Multi-model (Miyazaki et al., 2019b), Multi-resolution (Sekiya et al., to be submitted)
- **Other activities:** TROPOMI multi-constituent, AQ-GHG synergies, VOCs, Higher resolution (GCHP)





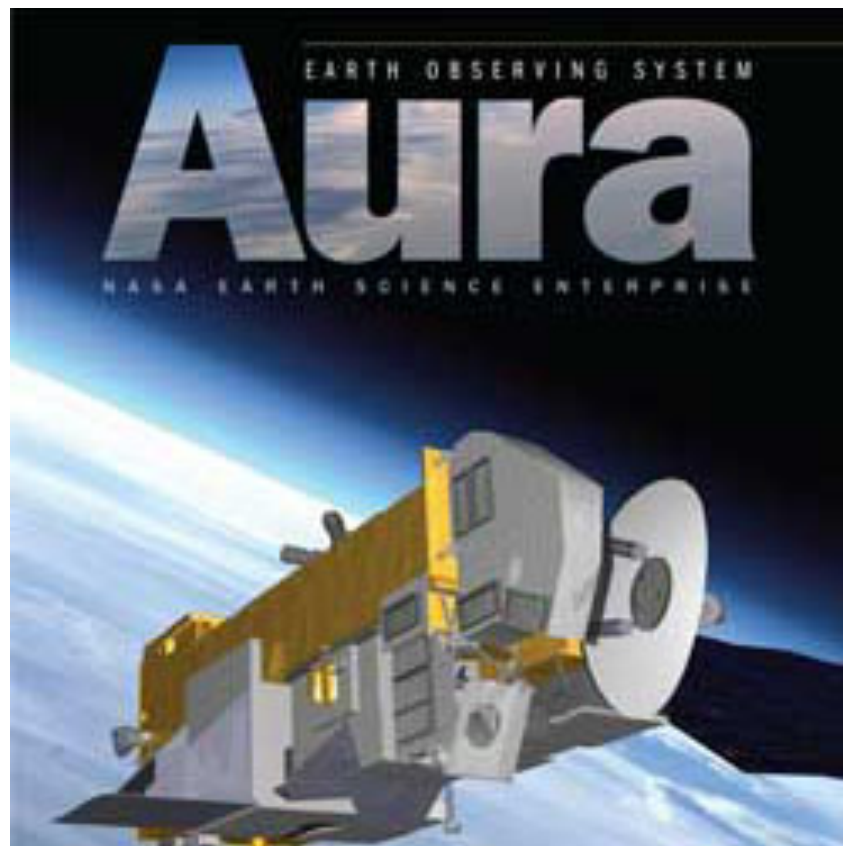
# Conclusions

- A 14-year chemistry reanalysis has been conducted using multi-constituent multi-sensor satellite data assimilation, in order to provide comprehensive information on atmospheric composition and emissions variability.
- The reanalysis data, combined with suborbital and ground-based measurements, has been used to improve our understanding of atmospheric composition and their impacts on air quality, human health, ecosystem, and climate.
- Diagnostic information readily available from MOMO-Chem has the potential to improve chemical predictions through relationships such as emergent constraints.

## **Relevant ongoing work at JPL**

- Evaluations of new satellite data (AIRS/OMI, TES/OMI, GOME-2/IASI)
- AQ-GHG synergy: multi-species constraints and joint emission estimation (OCO-2,3)
- Emergent constraints on the chemistry-climate system and carbon cycle (CMS-Flux)
- Health impact assessment (NASA HAQAST)



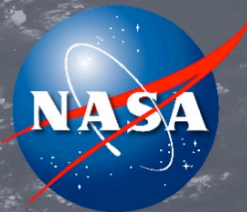


**Thank you!**

**You made  
chemical reanalyses**

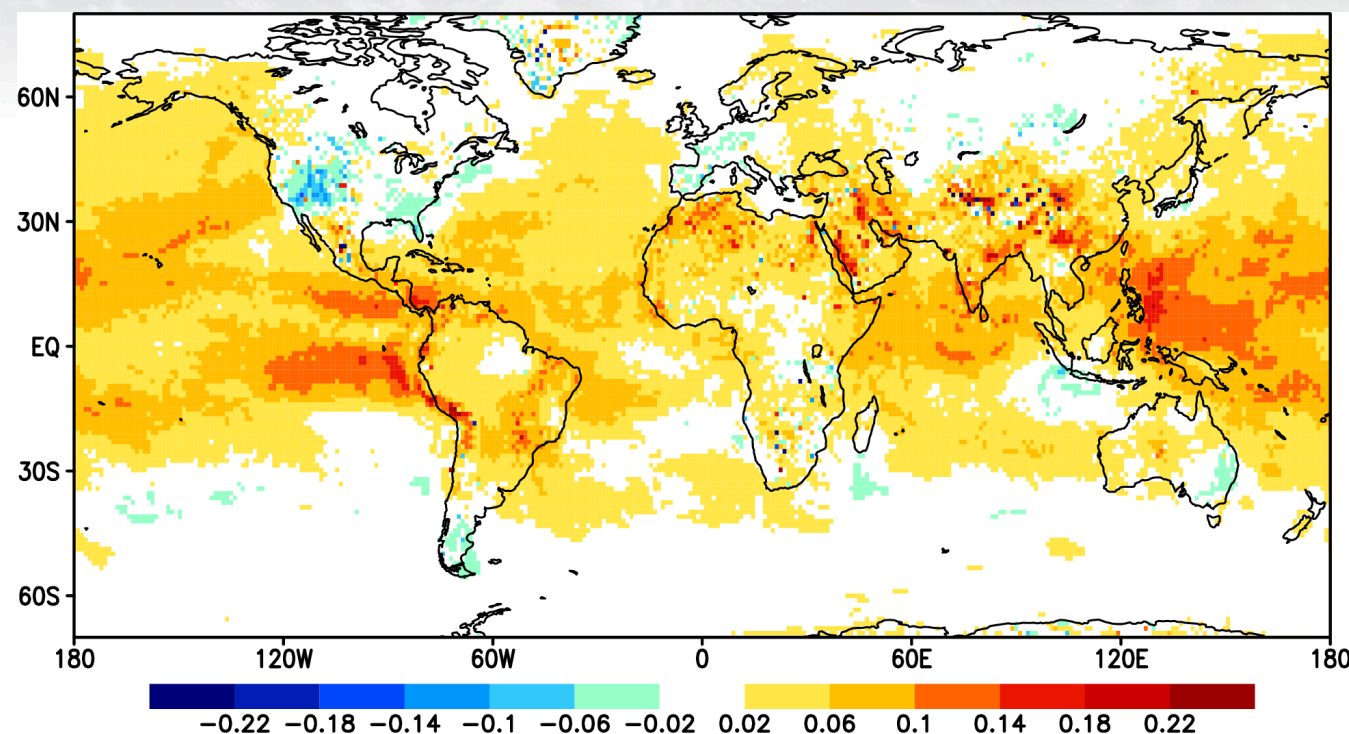
backup slides



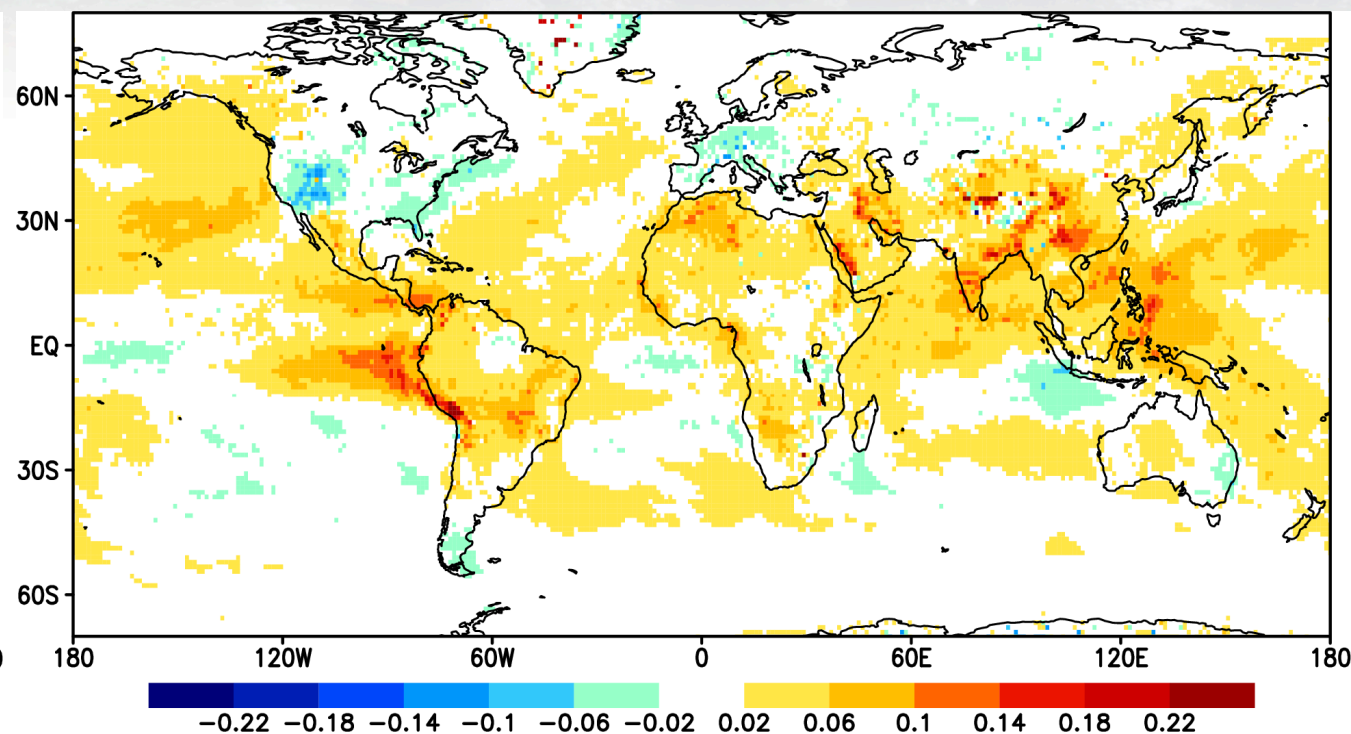


# Attribution of changes in oxidizing capacity

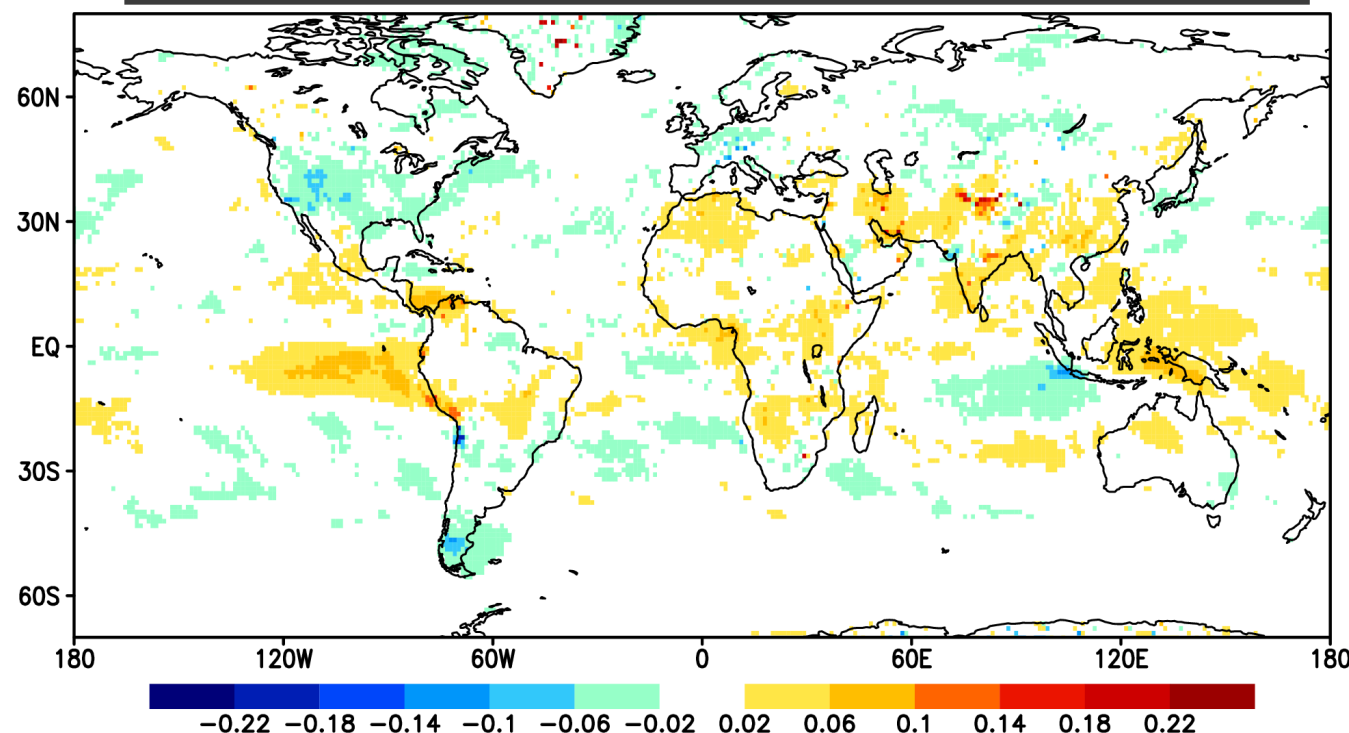
## Reanalysis (w/o TES)



## due to NOx emission changes



## due to meteorology

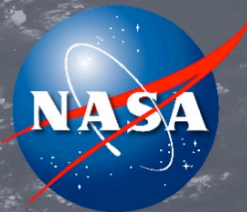


Tropo. OH changes :  
2010-2016 minus 2005-2009

### For improved pollution exposure assessment

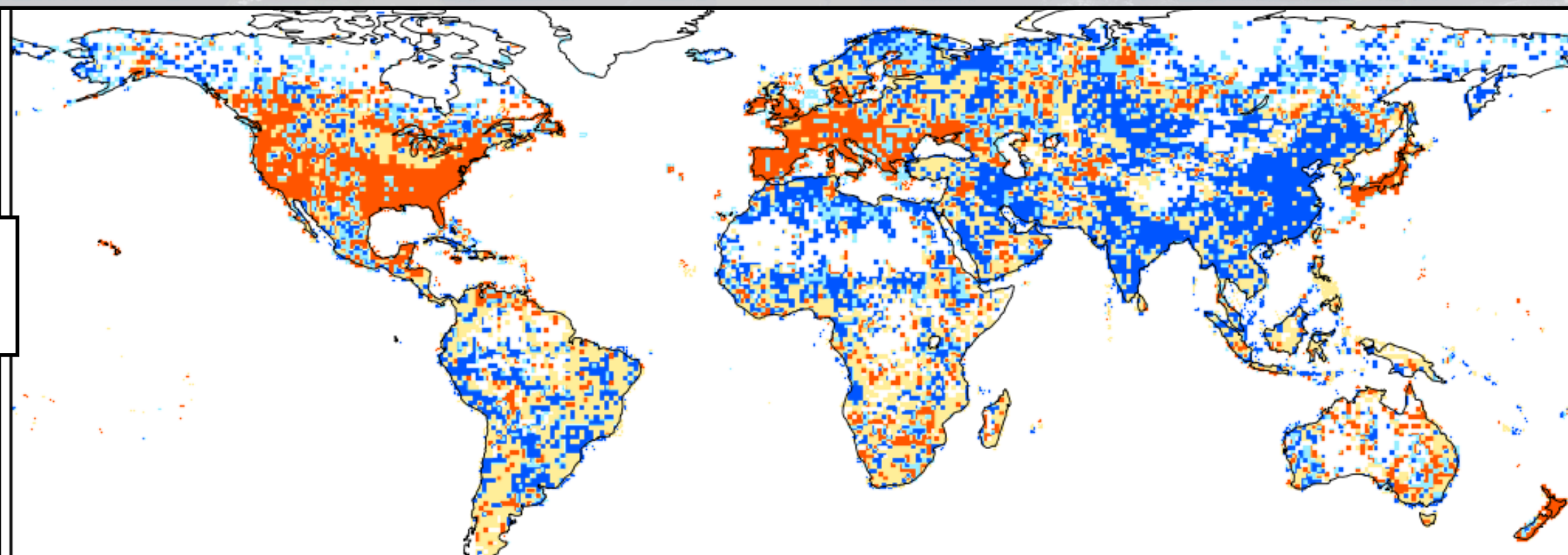
- The limited spatial coverage of this network is clearly insufficient for global health impact assessment especially in developing countries that suffer from severe



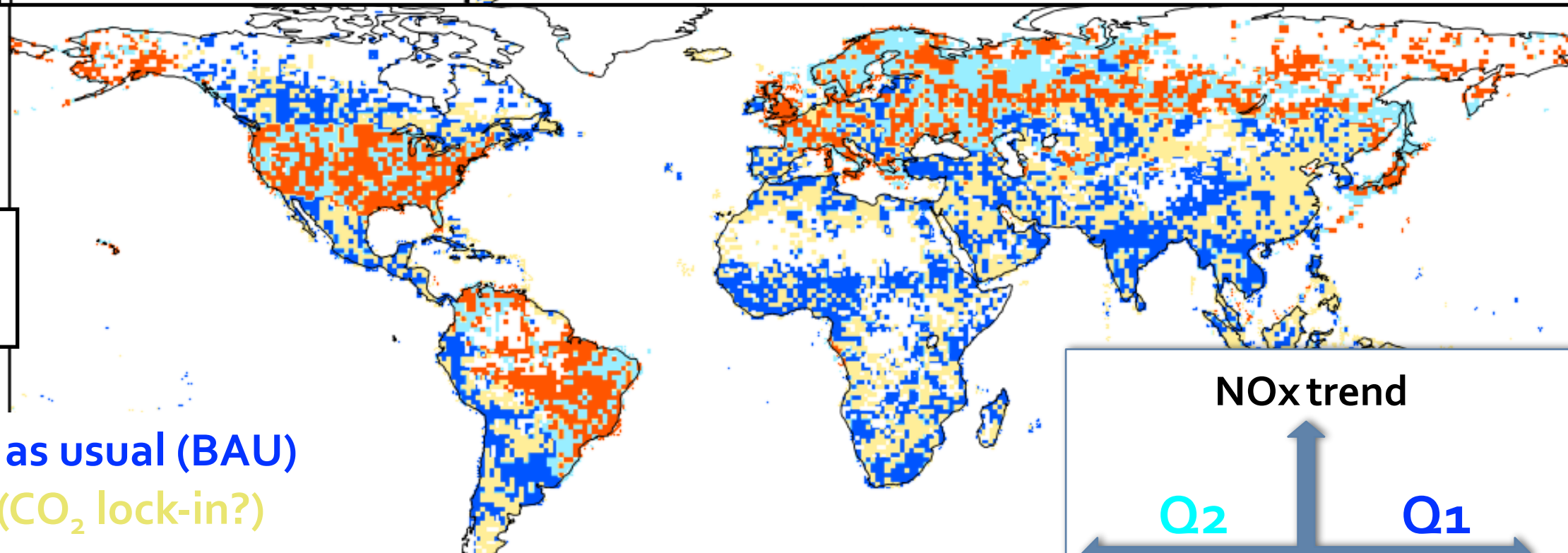


# Carbon/Air quality co-evolutions

2005-  
2010



2011-  
2017



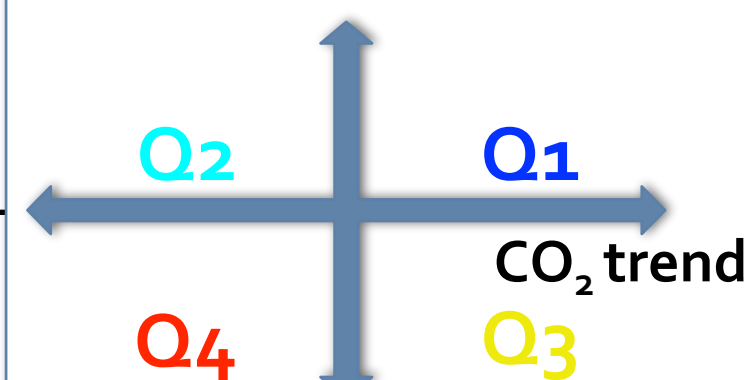
Q1: Business as usual (BAU)

Q3: AQ-only (CO<sub>2</sub> lock-in?)

Q4: AQ/Carbon (renewables)

Q2: Carbon-only

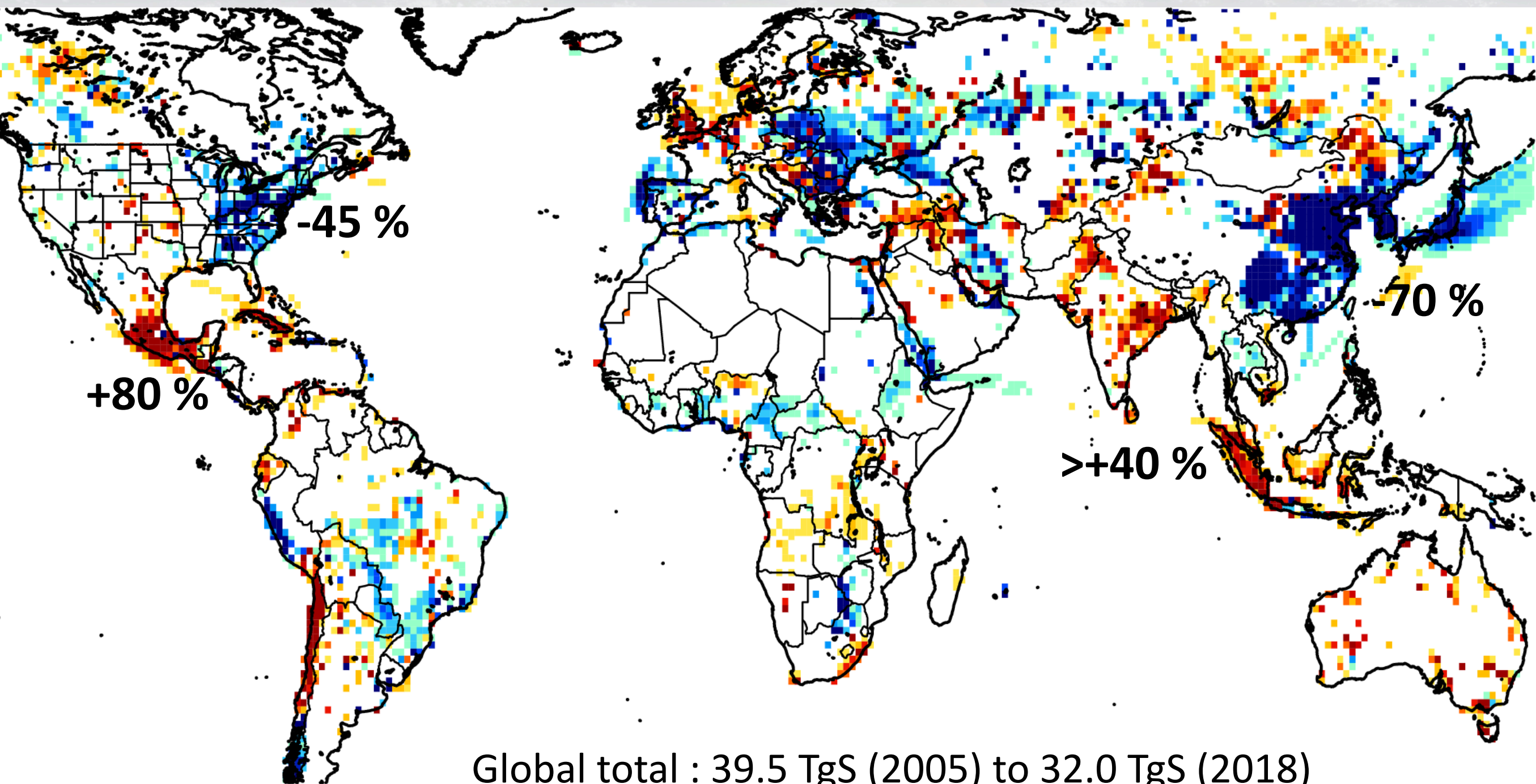
NO<sub>x</sub> trend







# Global SO<sub>2</sub> emission trends (2005-2018)



Global total : 39.5 TgS (2005) to 32.0 TgS (2018)

→ *Aerosols, climate, human health*





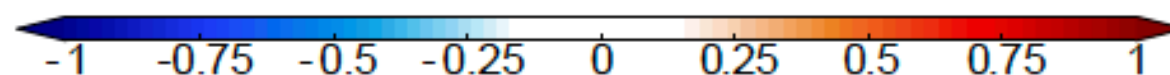
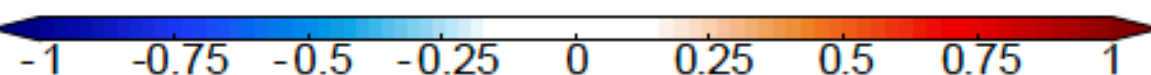
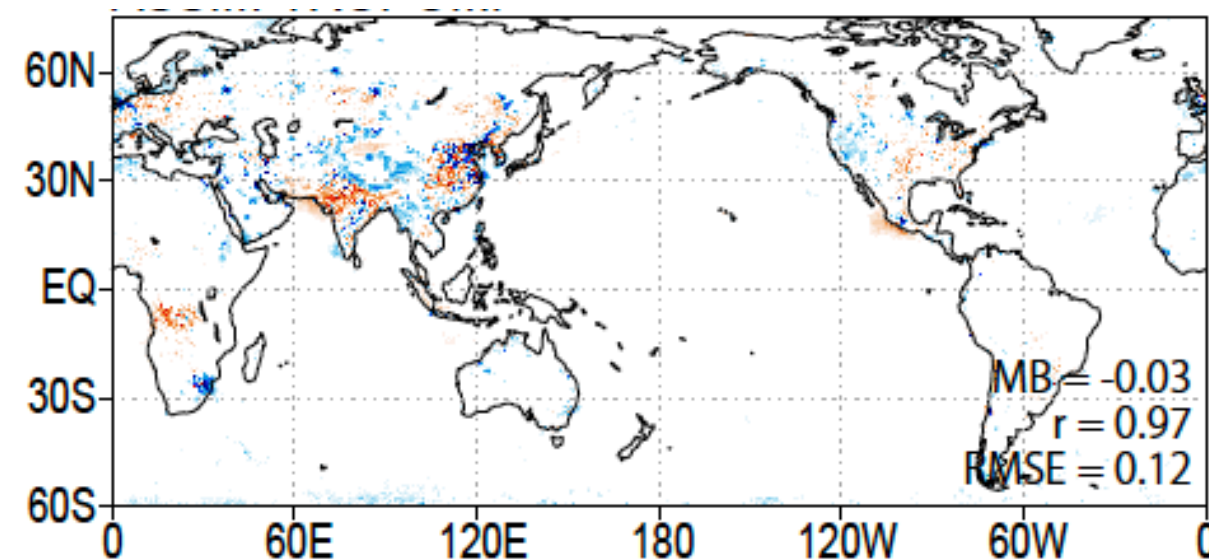
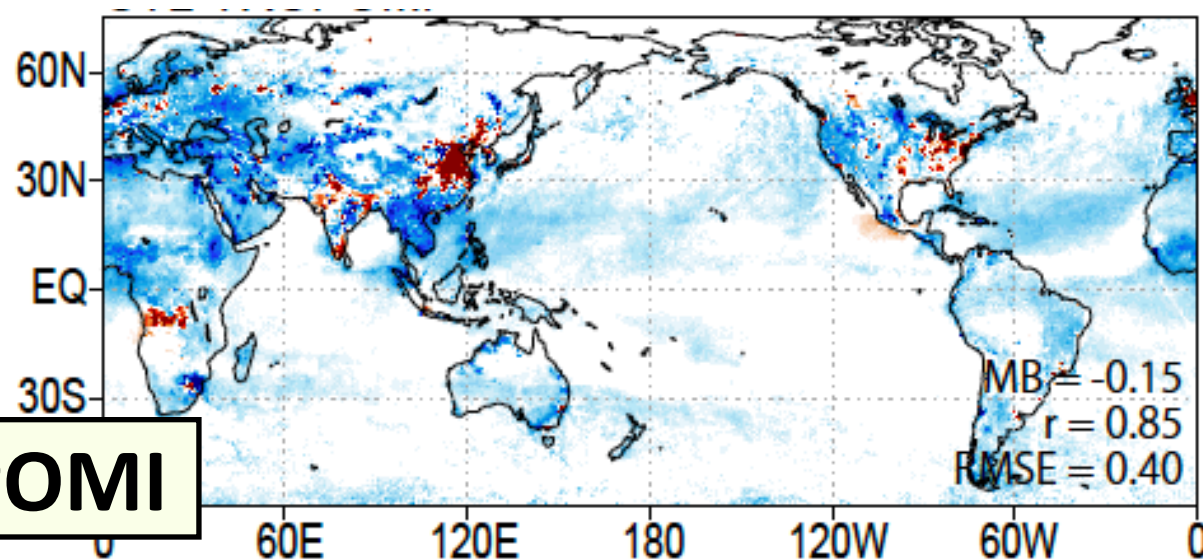
# For improved evaluation of emission changes

Global 0.5° resolution assimilations of TROPOMI NO<sub>2</sub> (May 2018)

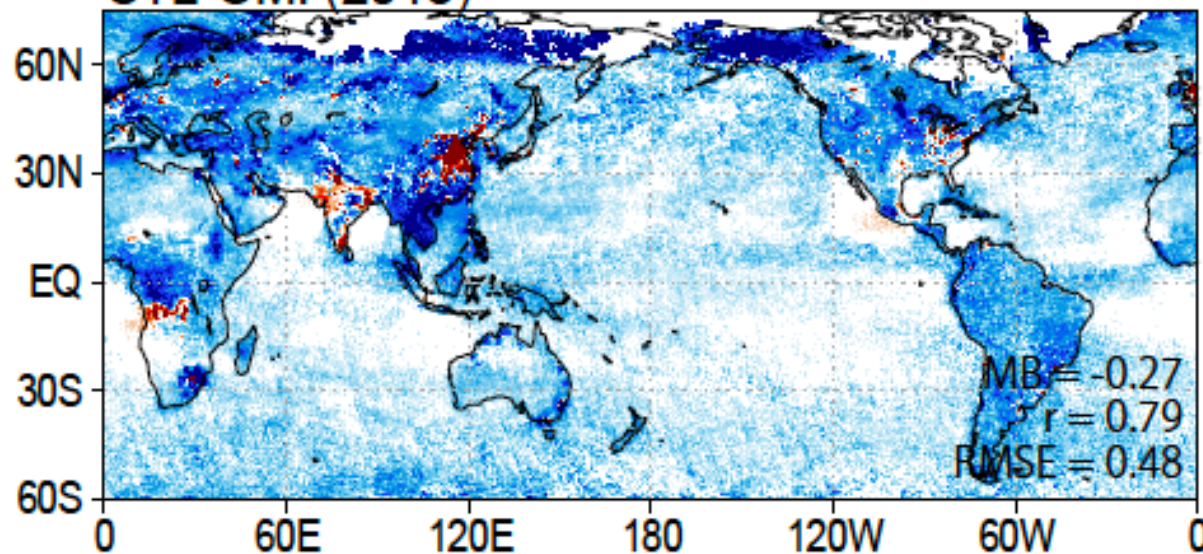
**Model-Obs**

**Assimilation-Obs**

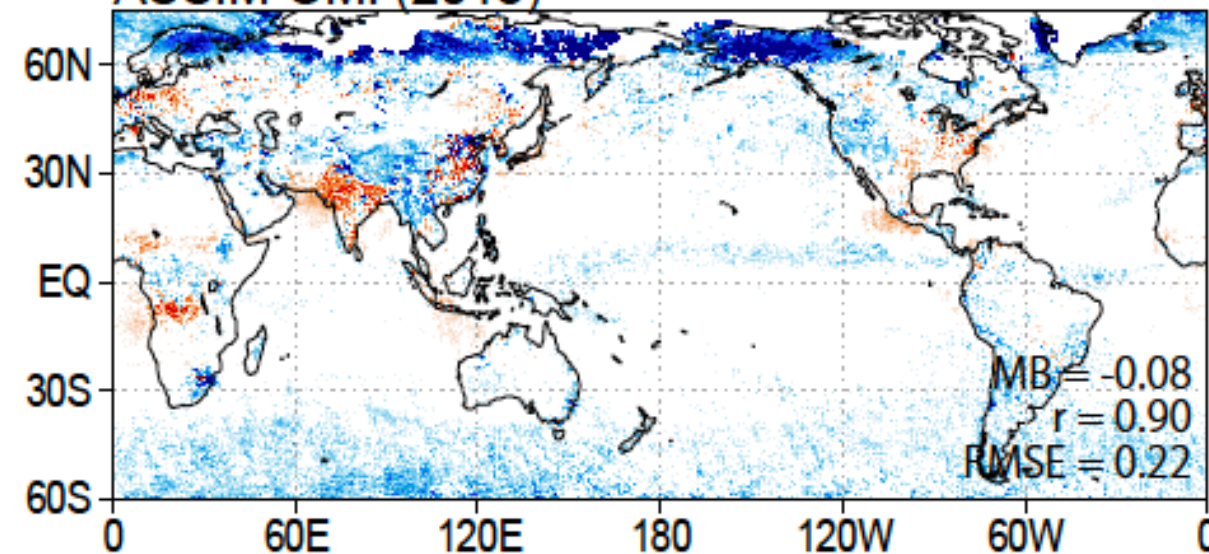
**TROPOMI**



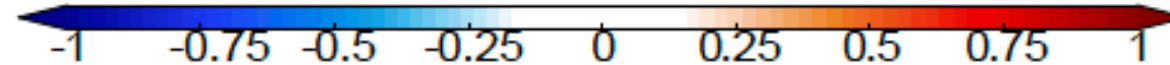
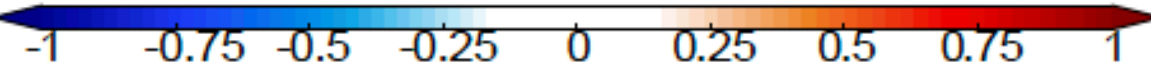
**CTL-OMI (2018)**



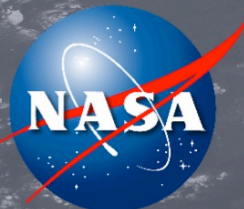
**ASSIM-OMI (2018)**



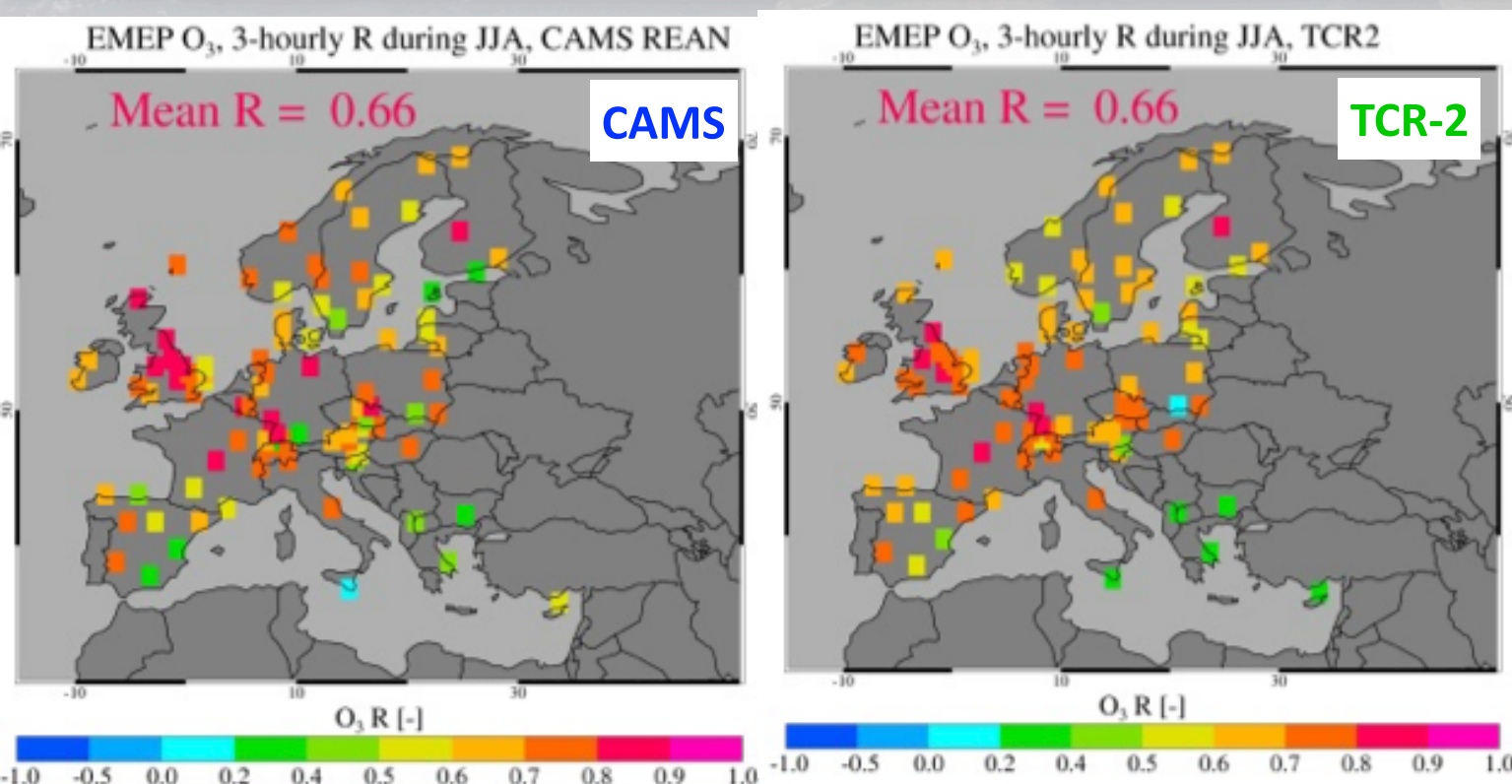
**OMI**



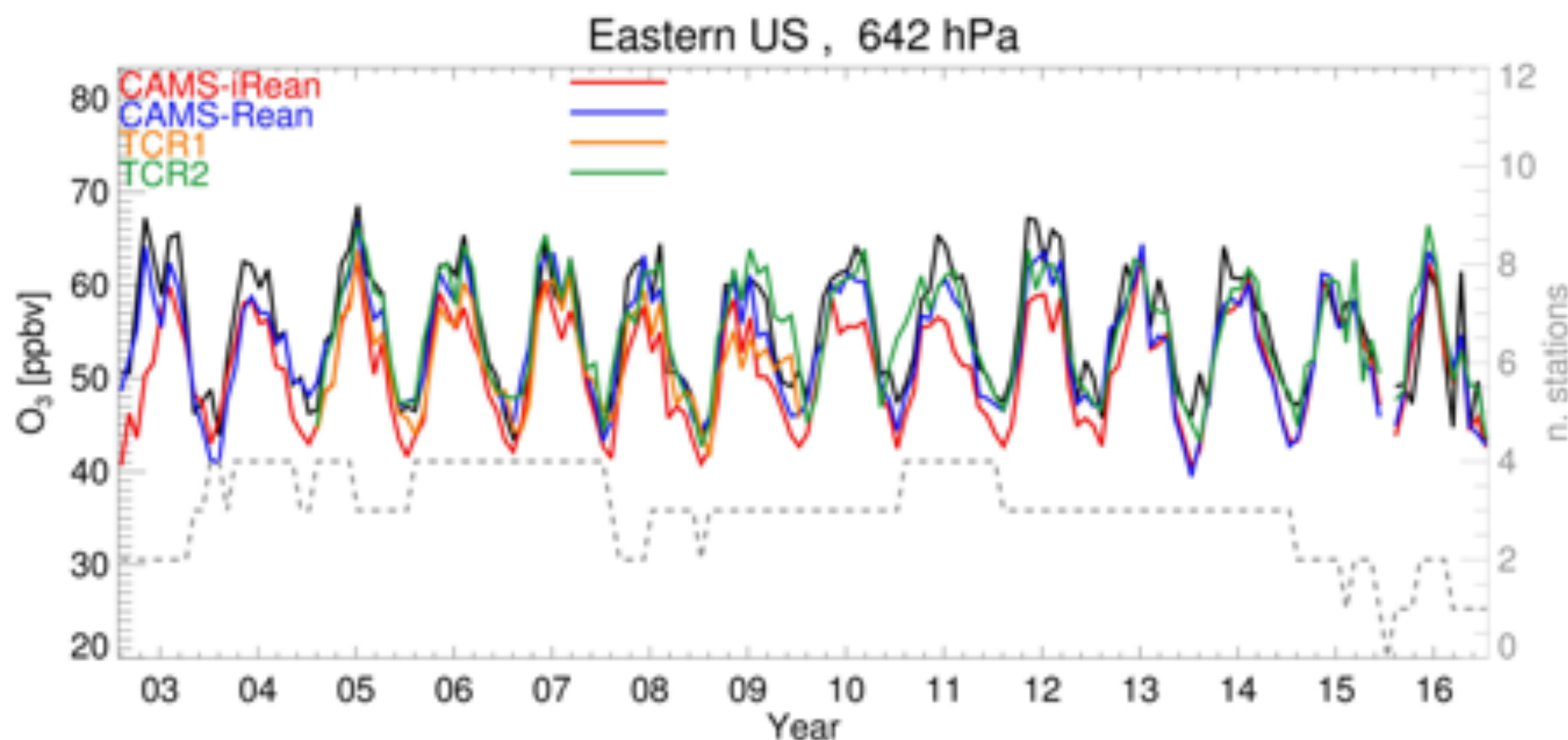




# Ozone reanalysis inter-comparisons



Products	Model	DA	Period
CAMS-iRA	IFS (CB05) T159 (1.1)	4D-VAR	2003-2018
CAMS-RA	IFS(CB05)+Aerosol T255 (0.7)	4D-VAR	2003-present
TCR-1	CHASER-EnKF T42 (2.8)	EnKF	2005-2016
TCR-2	MIROC-Chem-EnKF T106 (1.1)	EnKF	2005-2018



## RMSE (ppbv)

CAMS-iRA	4.9
CAMS-RA	3.2
TCR-1	5.0
TCR-2	3.4

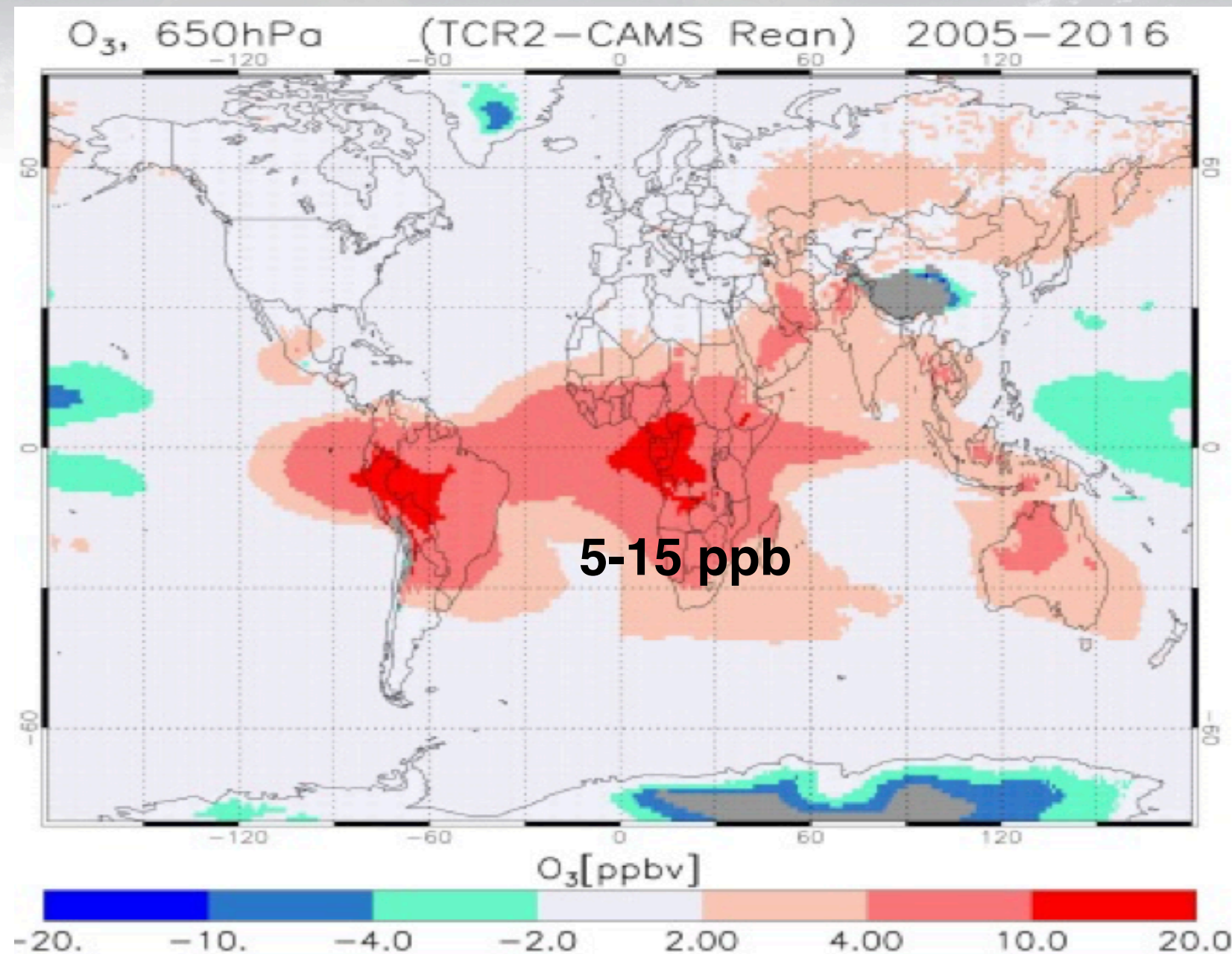
Huijnen et al., submitted





# Ozone reanalysis inter-comparisons

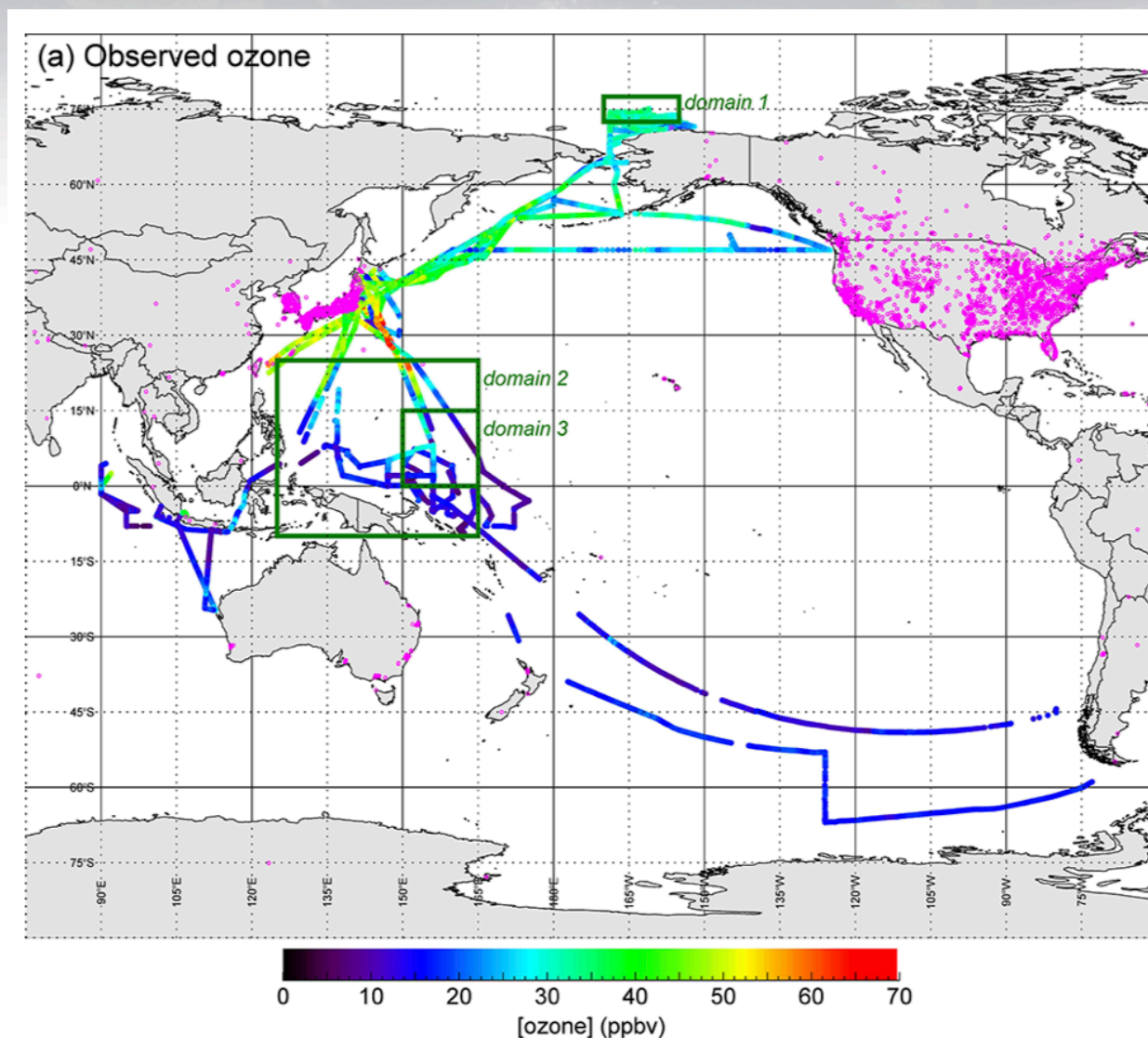
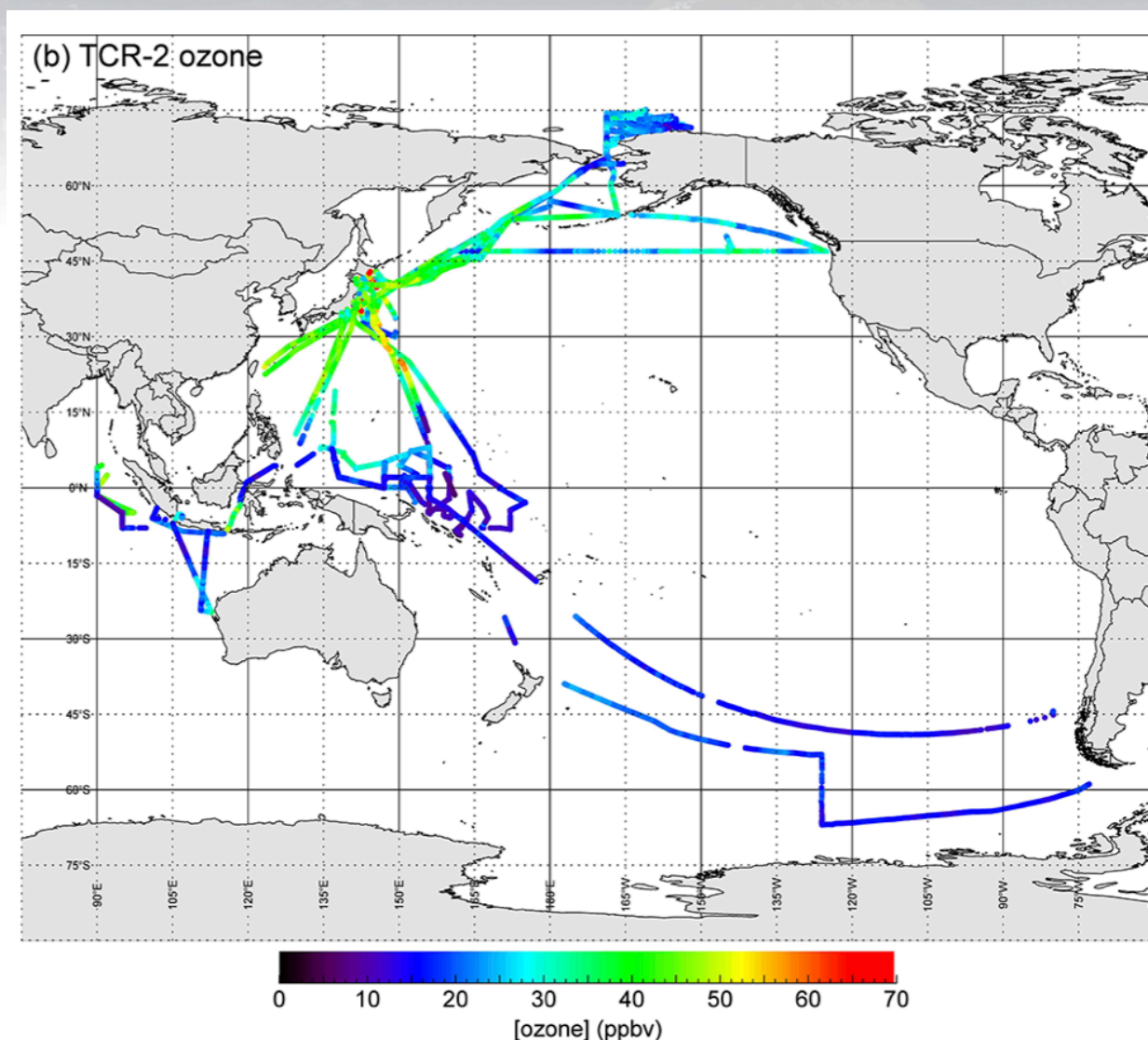
TCR-2  
minus  
CAMS



Over the tropical lands, the representation of biomass burning emissions, biogenic emissions, their impacts on ozone production, as well as convective transport could be largely different among the systems.



# Surface ozone reanalysis over remote oceans



- TCR-2 reproduced well ( $R=0.78$ ) photochemical buildup of ozone and enabled interpretation of observations regarding pollution sources.
- Tropical western Pacific ozone remains a challenge but can be improved by the incorporation of halogen chemistry and additional observations.
- The ability to reproduce remote ocean ozone observations demonstrates that chemical reanalyses can be used to help estimate the global mean OH.